

## LA-UR-15-24386

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Title: Materials Frontiers to Empower Quantum Computing

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Intended for: Workshop report  
Report

Issued: 2015-06-11

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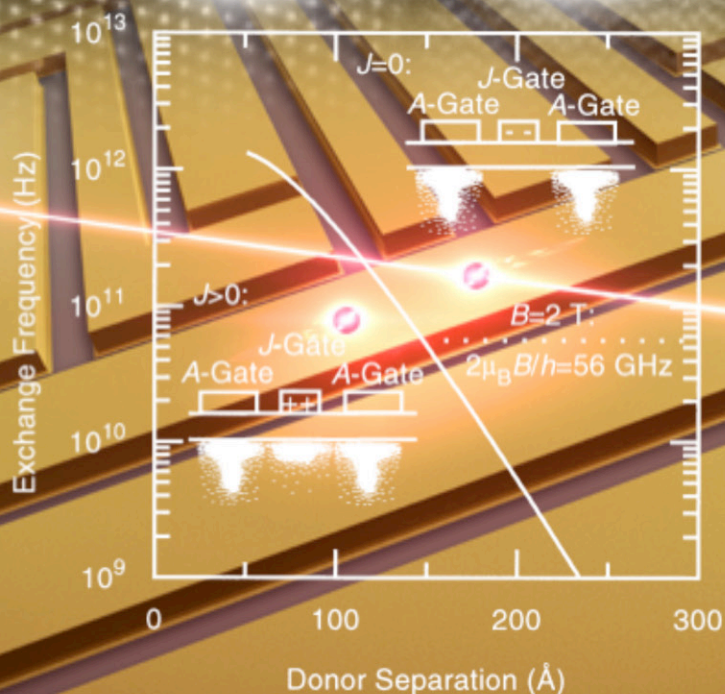
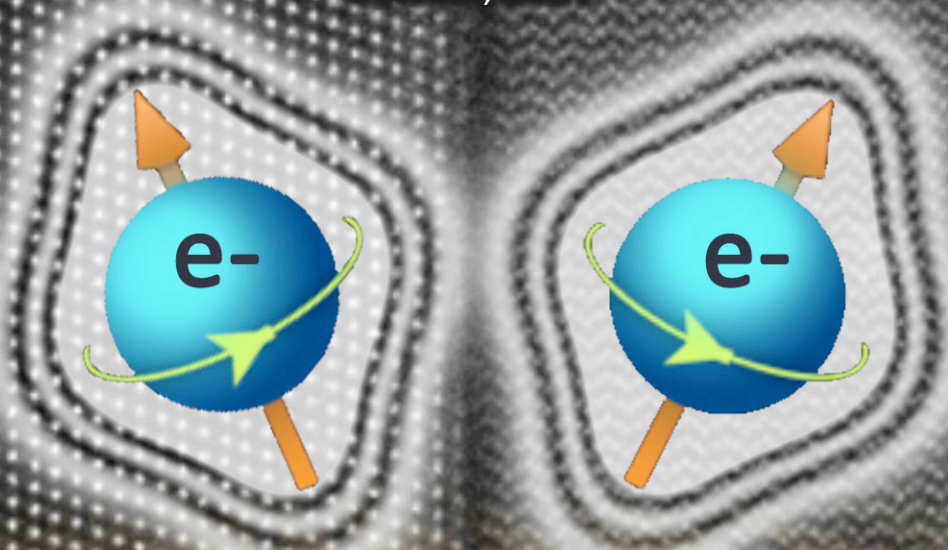
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# Materials Frontiers to Empower Quantum Computing

A Report of a Workshop on  
Materials Opportunities for Quantum Computing

Held in Los Alamos, New Mexico

October, 2014







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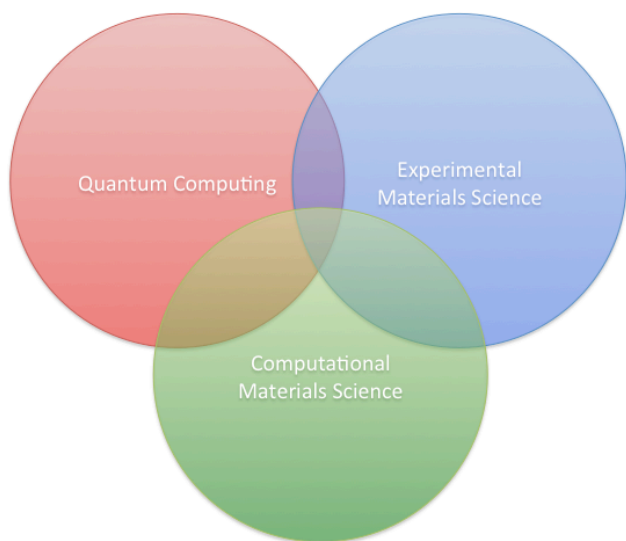
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## Executive Summary

This is an exciting time at the nexus of quantum computing and materials research. This recognition was the impetus for convening a workshop of approximately 75 leading researchers spanning the fields of quantum computing and materials science and also spanning academia and the U.S. Department of Energy (DOE) national laboratory system. The workshop unambiguously succeeded in bringing together diverse communities, nucleating new collaborations, and perhaps most importantly, identifying new research frontiers. This report captures those insights, summarizes the current state of research and identifies future directions in computational and experimental sciences that will extend the deliberate creation and control of individual quantum states of matter, a grand challenge at the frontiers of both quantum computing and materials research.



*Researchers from three scientific specialties gathered together to nucleate new collaborations and identify new research frontiers in the emerging interdisciplinary area of materials science challenges related to quantum computing (Richardson 2014).*

This is an exciting time because quantum computing is at a turning point—from “if” various qubit technologies are possible towards the predictive enhancement of their performance beyond serendipity. Heroic efforts by single investigators and small groups have laid a strong foundation. The present opportunity is to systematize and accelerate these gains through strategic teaming.

This is also an exciting time because materials research is at a turning point—making a transition from observation of properties to prediction and control of functionality. The current states of the art of large-scale simulation, high-resolution characterization, and exquisite control of materials synthesis are simultaneously on the cusp of robustly cooperating in a fashion that can dramatically impact our knowledge of how individual quantum states interact with matter.

The materials frontiers described in this report represent significant advances in electronic materials and our understanding of the interactions between the local material and manufactured quantum states. Simultaneously, directed efforts to solve materials issues related to quantum computing provide opportunities to control and probe the fundamental arrangement of matter that will impact all electronic materials. Exciting opportunities to harness quantum devices as materials characterization tools can provide valuable insight that can be used to propel materials science research.

Limitations in qubit performance can be traced back to disorder and impurities in the constituent materials from which they are made as well as to sources of noise and decoherence that arise from the qubit's coupling to its environment and associated defects. This motivates the refinement of materials and drives the need for greater synthesis fidelity. Translating “quantum-grade” materials into reliable qubit devices requires fundamental improvements in existing fabrication tools and techniques. Successfully fabricating quantum devices, and authenticating the approach with integrated models to enable a predictive capability for “quantum-grade” fabrication and processing, offers potentially dramatic improvements for quantum computing and electronic devices in general. The need for mesoscale techniques that will allow device-scale models and measurement analysis approaches to leverage first principles atomic-scale simulations is also clear. The investigation of the absolute quantum ground state and near-ground states through equilibrium studies and dynamical interrogation poses challenges to existing techniques and expertise. Leveraging existing DOE user facilities to precisely characterize material chemistry and structure is a critical first step to understanding the electronic structures of “quantum-grade” materials.

Given the magnitude of the challenge, integrated team efforts with expertise that spans quantum computing and materials research are necessary for success. In so doing, with balanced advances in simulation, characterization, and synthesis, there is also the opportunity to grow a generation of interdisciplinary researchers equally fluent in qubit physics and materials research approaches. Similarly, significant materials science capabilities exist, especially at the DOE laboratories that can be brought to bear on the identified challenges. These initial efforts will not only accelerate progress but also motivate the development of even more advanced capabilities as the challenges become clearer.

In the end, workshop participants strongly agreed with the framing premise that this is an exciting time at the nexus of quantum computing and materials research. This report attempts to capture their enthusiasm. In what follows, the current state of the art in quantum computing, spanning four different qubit technologies, is summarized; overarching grand challenges and actionable research opportunities are identified; and suggestions for meeting these opportunities are proposed.



# Introduction

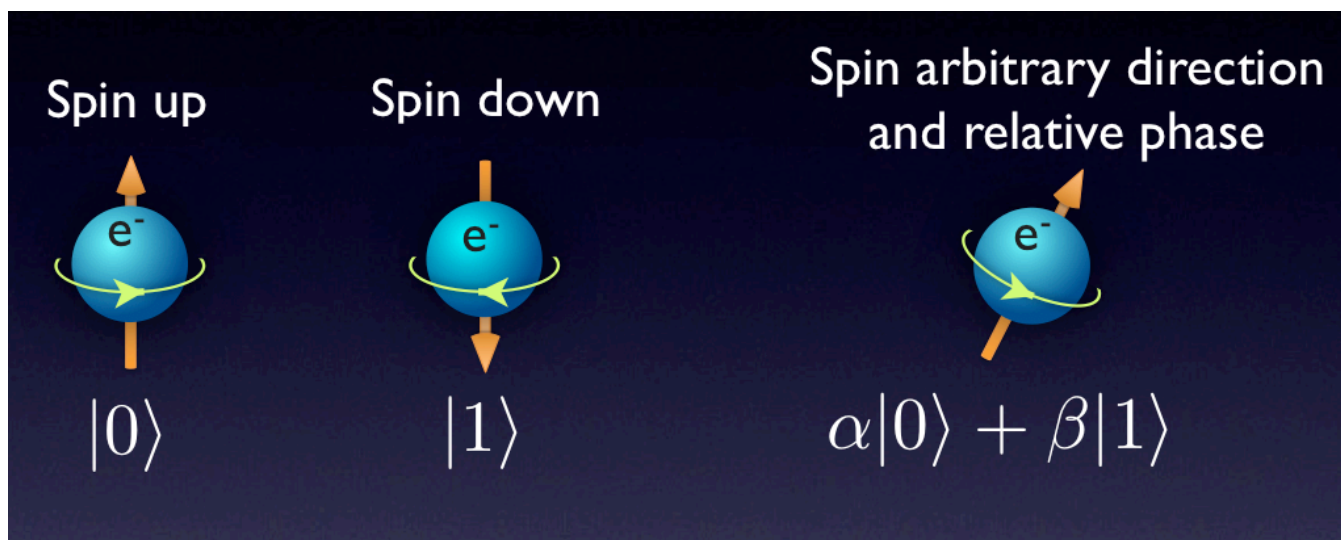
## Motivation

Quantum computing promises to revolutionize information technology as we know it today. By changing the physical nature of the bit, the fundamental unit of information, entirely new frontiers of computational mathematics are feasible. These newly enabled calculations will function on an entirely new computer architecture that is expected to radically change how quickly some computationally difficult problems are solved. The physical limitations to creating a quantum computer originate in our ability to manipulate and control the basic materials that comprise quantum computing devices.

Research in the area of quantum information technology is seeking to answer several questions, such as whether a specific mathematical function can be answered or computed, and if so, what is the cost of computing the answer. Here, “cost” refers to the computational complexity; i.e., the physical resources such as the number of logical operations, computation time, required memory, and energy that must be invested to solve the problem of interest. Computing problems are classified based on how the cost scales, as either exponential or polynomial scaling with respect to the initial size of the problem. En route to addressing these questions, computer scientists compare the implementation of various algorithms in both classical computing architectures and

quantum computing architectures, which, for example, include quantum gate arrays, adiabatic computing based on quantum annealing, and topological approaches using braiding of anyons. The requirement to perform calculations on a potential computational architecture provides specific criteria for the underlying quantum system. Although the frontiers of quantum algorithm development were not within the purview of the present workshop, there is broad recognition of the benefit that can result from building synergy and integration between the quantum materials, quantum computing architectures, and quantum algorithms communities.

The qubit is the quantum analogue to the bit. In classical digital computing, the bit is a voltage defined by the system designer to represent either a 0 or a 1. Transistors are then arranged in numerous configurations to deterministically perform desired operations on the input bits. Qubits, on the other hand, are defined by a quantum mechanical two-level system, a continuous rather than a discrete state space, that results from placing a quantum entity (e.g., Cooper pair, electron, ion, or flux quanta) in an engineered environment that creates a particular energy landscape. Therefore, the specifics of a qubit state are intimately linked to the qubit architecture and its environment. Furthermore, the engineering design decisions associated with a qubit state cannot be easily adjusted.



*Quantum computers achieve their computing power from the physics governing qubits, represented here as spin states, that can exist in superposition states that are entangled with unlimited number of other qubits. (Eriksson 2014)*

Qubits can exist in linear combinations of quantum states. That is, if two states are defined as the computational basis  $|0\rangle$  and  $|1\rangle$ , then the qubit can exist in the state  $\Psi = \alpha|0\rangle + \beta|1\rangle$ , where  $\alpha$  and  $\beta$  are complex numbers. Additionally, multiple qubits can be intentionally entangled which means that the qubits cannot be described independently even though they may be physically separated. Therefore, the exact *classical* specification of a qubit state requires an infinite amount of information to be stored by the qubit. The problem is compounded by complex qubit-qubit interactions in the system. Unfortunately, when information is read out of the qubit system, the associated measurement projects (or “collapses”) the quantum information into a classical output. Therefore, the effect of measurements on the quantum state is inescapable, and much of the information stored in the quantum state and its interactions are hidden from direct observation. However, it is these hidden quantum effects that are exploited by quantum algorithms to provide substantial computational efficiency increases for certain classes of computations.

All quantum computing technologies require that qubits be able to be reliably initialized into a predefined state, manipulated to exchange information between qubits in a fully quantum mechanical manner, and precisely measured to determine the result of the quantum interactions. The intimate connection between the quantum architecture, the qubit, and the surrounding environment imposes significant demands on quantum computing technologies and the materials that underpin them.

In recognition that direct assignment of the two quantum states is not really possible, but rather engineered through the local environment, the material challenges therefore result from indirect control and the inadvertent introduction of other states or excitations that can disturb the ideal qubit states. The relative difficulty in adjusting these effects depends on the specific system, and in fact, the need to learn how to better manipulate and control these effects is the point of this report.

In other words, systems that use quantum devices as the fundamental unit of information require that the surrounding environment and design features be constructed with precision that limits variability so that its imperfections result in distortions to the local environment that are smaller than the characteristic states that are used to store information. Understanding and controlling the electronic, magnetic, and lattice environment of the qubit at a sufficient level to store and manipulate quantum information is indeed a grand challenge.

In recognition of these new technological demands on materials, this report introduces the concept of quantum-grade materials. Currently, materials scientists and practitioners are familiar with solvent-grade, mechanical-grade, and electronic-grade materials as distinct levels of purity with impurities often measured in parts per trillion. Success in electronic-grade materials is reflected in our ability to manipulate semiconductors to great effect in modern electronic devices. However, the quantum computing requirement of maintaining the quantum mechanical features of the qubit imposes purity and structure requirements that exceed those of current electronic-grade materials. Progress in quantum computing research is currently limited by the gap between our electronic-grade and quantum-grade understanding of materials science. Advancing our materials understanding from observation and validation to prediction and control of quantum-grade materials can bridge this gap.

As the state of the art is pushed simultaneously in modeling, characterizing, and fabricating materials of technological importance to quantum computing, it is expected that unanticipated discoveries will be made as the precision of these disciplines are refined and improved beyond the current state of the art. Mastery of quantum-grade

materials will likely also enable a new generation of computing technology and characterization techniques. By challenging our most accurate models, measurement tools, and synthesis systems, the opportunities outlined in this report hold promise not only for improving quantum computing technology but also hold promise for advancing the frontiers of materials research more broadly.

## **Primary Quantum Computing Technologies**

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The experimental realization of a working quantum computer is currently beyond our scientific and technological ability to predict and control materials. Experimental research in numerous quantum computing technologies has matured over the past decade. Much of the progress has been the result of isolating a few qubits from the impurities and disorder inherent in the technologically limited qubit materials. The most promising qubits today are made from silicon, superconducting circuits, and individual ions. The ability to manipulate these qubits has been demonstrated, at least by a few groups.

### ***Silicon-based Quantum Computing***

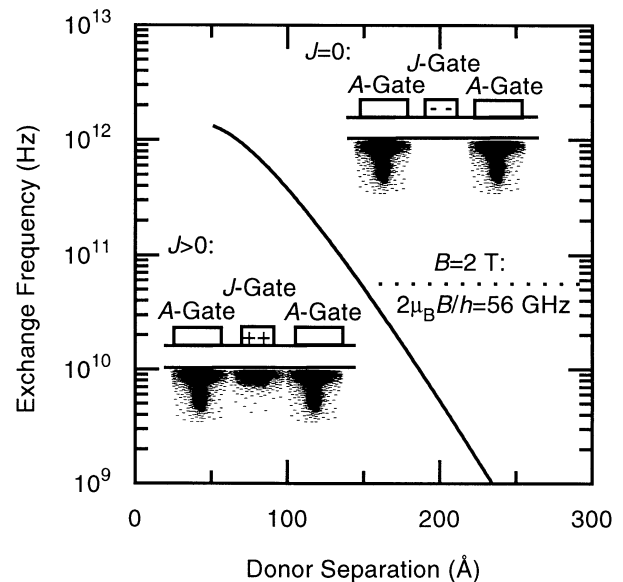
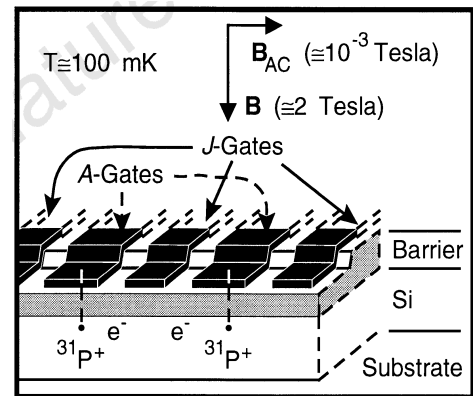
The semiconductor industry has invested an estimated \$3T developing sophisticated capabilities to fabricate microelectronic devices including today's classical computers. Silicon quantum computing devices promise to leverage the investments and advances made by the microelectronics industry. This synergy allows advances from the larger semiconductor industry to accelerate the development of qubits. Conversely, the synergy also means that breakthroughs in silicon quantum computing devices can help define and drive development of post-CMOS devices. It is worth noting, however, that the physical and computational bases for silicon

quantum computing differ substantially from those used in classical computing devices. The quantum effects being confronted in classical computing devices as they continue to shrink are only one aspect of the physics used in silicon qubits.

Silicon-based qubits are defined using two- or few-level systems, but they are in practice made from systems with large numbers of states that can inadvertently cause transitions to states outside of the computational basis. Such transitions represent errors resulting in qubit information leakage. Both electrons and spin- $\frac{1}{2}$  nuclei naturally form two-level systems in a magnetic field, which can form usable quantum computational bases. Both spin and charge qubits are used in quantum computing devices; however, the presence of charge disorder and noise in silicon devices generally limits the usefulness of charge qubits in silicon.

Two primary methods are used to define silicon qubits: donor atoms and electrostatically gated quantum dots. Donor atoms are n-type dopants from group-V elements such as phosphorous or arsenic that contain an extra positive charge and electron compared to Si. They form substitutional defects in the Si lattice, and the extra nuclear charge and electron form a hydrogen-atom-like addition to the Si band structure that can be manipulated as a qubit. The use of donor atoms in silicon as a platform for quantum computing dates to the work by Kane in 1998<sup>1</sup>. Donor qubits typically use electron spin to define the computational basis: opposing spin orientations with respect to an external magnetic field are used to define the  $|0\rangle$  and  $|1\rangle$  states. Single-qubit gates may be applied using an external electron spin resonance (ESR) field. Electrodes over the donor atoms can be used to Stark-shift the electronic wavefunction on or off the atom, and thus in and out of resonance with the

ESR field. The change in resonance with the ESR field modifies the global ESR interactions to affect local interactions on selected atoms. Exchange interactions between electrons on adjacent donors, often modulated using another surface gate between the adjacent donors, can be used to create logical (2-qubit) gates. Qubit readout may be performed with spin-to-charge conversion using spin-dependent tunneling into a readout single-electron transistor.



*Illustration of the Kane quantum computing architecture containing phosphorous donors and electrons in a silicon host, separated by a barrier from metal gates on the surface. Gates vary the electrostatic potential barrier between donors to enhance or reduce exchange coupling, proportional to the electron wavefunction overlap. (Kane 1998)*



Donor atoms are normally implanted into Si substrates using ion beams similar to those used for implants in standard fabrication techniques. Such techniques require the donor atoms to be fired at the substrate at a relatively high energy of 1-10s keV. This process results in a probabilistic placement of the donor atoms within some “straggle” distance about the intended position. For systems of relevance, the straggle is often of the same magnitude as the intended separation of the donor atoms (e.g., 50 nm), thus causing substantial variability in the resulting device parameters. Some techniques have been developed to circumvent this issue. Specifically, demonstrations of placing donor atoms with atomic precision on a Si surface have been completed with scanning tunneling microscopy (STM)<sup>2</sup>. Clark, Simmons, and coworkers have developed the application of STM lithography for the development of Si donor qubits<sup>3</sup>.

The majority of donor atoms have nuclear spins, which presents the additional possibility of using the nuclear spin as a qubit in donor atom systems. Nuclear spins have substantially longer coherence times than electron spins, particularly for ionized nuclei. However, nuclear spin systems also interact more slowly with external fields, which means that single-qubit gates using nuclear magnetic resonance rather than ESR are slower, and other interactions are more challenging to define and control.

Silicon quantum dots, where an electron in a 3-D potential well defines the qubit, provide an alternative to donors for silicon-based qubits. Both charge-based qubits and spin-based qubits have been demonstrated, although spin qubits are typically seen as more promising for quantum computing applications. Much of the progress in this field derives from early work on GaAs/AlGaAs materials that show significant

promise in defining sophisticated qubits and related structures. Unfortunately, GaAs-based systems suffer from short qubit lifetimes that are limited by Overhauser fields produced by the nuclear spins of the GaAs material itself. The move from GaAs to Si was motivated by the potential to develop similar devices in a nuclear-spin-free environment using isotopically enriched Si. Quantum dots use doping, electrostatic gating, or Si/SiGe heterostructures grown on relaxed SiGe buffers to form a two-dimensional electron gas at the Si/insulator interface or in a buried Si quantum well. All of these implementations use additional electrostatic gates to confine and shape the local potential of the two-dimensional electron gas to create few-electron puddles that comprise the qubits.

As in the donor systems, one can use electron spins to define qubits. However, the added flexibility allowed by using electrodes to define arbitrary potentials has enabled other methods of encoding qubits. Promising encodings include using the singlet and triplet states of a pair of electron spins to define the computational basis, as well as using low-lying states of 3-qubit systems to define robust qubits.

The same physics that allows fundamentally greater computational power to come from a few atoms makes these devices significantly more sensitive than traditional semiconducting devices to defects, disorder, and noise. The small scale of donor-atom and electrostatically gated quantum dot devices makes them sensitive to interfacial disorder on the same length scale. Additionally, the complex valley physics associated with the 6-fold degenerate conduction band minima in silicon is an undesired complication to silicon-based qubits. Moreover, the necessity for quantum devices to establish and

maintain entanglement makes these devices sensitive to environmental charge fluctuators that can render precise interactions difficult or impossible to control.

These challenges have slowed the rate of progress in designing high-fidelity silicon quantum devices. The challenges also necessitate the development of a complete silicon quantum device modeling capability tightly coupled to experimental characterization and device fabrication. This modeling capability needs to relate process parameters to the resulting quantum device characteristics to create quantum devices robust to defects, disorder, and noise.

Core silicon-based quantum technologies have reached a critical stage in their development: fundamental explorations have been performed, and two promising approaches (donors and dots) have been identified. Mastery of noise and disorder in these systems through advanced materials modeling and characterization techniques will be an important accelerator towards the realization of the potential of silicon qubits.

### ***Superconducting Qubits in Josephson Junction Circuits***

Following Leggett's seminal work on macroscopic quantum systems and the quantum measurement problem<sup>4</sup>, researchers have been able to demonstrate that Josephson junction circuits can be used to realize superconducting qubits<sup>5,6,7</sup>. These circuits exhibit quantum behavior with energy spectra largely determined by geometric parameters. With circuit quantum electrodynamics (cQED) guiding the development, several different types of superconducting qubits have been realized that quantize different entities such as charge, flux, or phase. cQED is a quantum field theory used to understand strong coherent coupling of artificial atoms with a

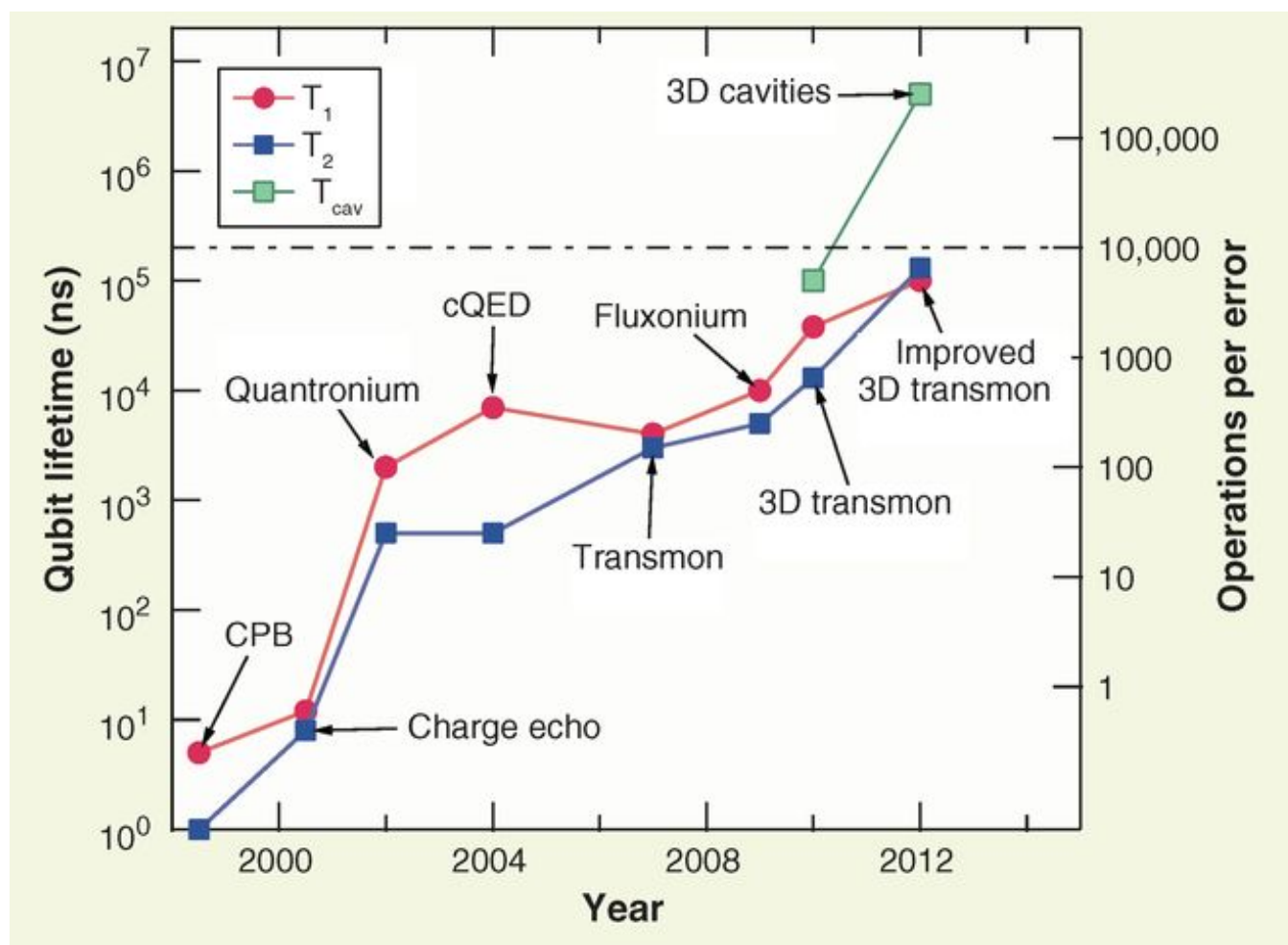
single mode of light. Superconducting qubits can be described as artificial atoms that are strongly and coherently coupled to a single mode of microwave radiation. In this approach, the microwave (i.e., light) mode is typically a standing wave in a superconducting (usually Al, but sometimes Nb or TiN) microwave transmission line or a resonant mode in a 3-D box<sup>8</sup>. The artificial atoms are typically constructed from mesoscopic-sized electrical circuit elements. In superconductor circuits, purely reactive linear circuit elements, inductors and capacitors, can be used to engineer evenly spaced energy levels of a harmonic oscillator. However, a non-linear circuit element must also be included so that a viable two-level quantum system with atom-like energy level spacing can be generated. The inherent non-linear inductance of a Josephson junction is used to provide the anharmonicity needed to generate quantum energy levels that can be individually addressed. Anharmonicity is important because it sets the limit on how short the control pulse signal can be to avoid mixing in higher energy levels beyond the two-level computational basis. It is therefore the linear and nonlinear circuit elements together that make superconducting qubits a potentially useful technology for realizing quantum computing.

A number of Josephson junction circuits have been used to make charge-, flux-, and phase-based qubits. To understand the differences in the various types of superconducting qubits, one needs to consider the relative ratio between the capacitive charging energy and the Josephson coupling energy. By choosing appropriate insulating material such as aluminum oxide and insulator thickness, the superconducting gap can be sufficiently large that only coherent Cooper-pairs tunnel across the junction.

When the appropriate gate voltage is applied to a charge qubit, only two charge states become relevant and the energy spectrum reduces to a two-state quantum system characteristic of a qubit. The transmon is a modified charge qubit that contains a Cooper-pair box (CPB): a mesoscopic device formed by coupling a small superconducting island to a superconducting reservoir through a Josephson junction. Consequently, the transmon is far less sensitive to charge noise, and hence more robust, than a standard charge qubit.

In a flux-based qubit, a mesoscopic-sized superconducting ring containing several Josephson junctions is engineered. When an external flux is applied to the loop, a persistent current is generated; i.e., Cooper pairs continuously flowing around the loop. The energy levels associated with the two different directions of the persistent current can be brought close together to form the qubit.

Presently, all types of superconducting qubits use aluminum and aluminum oxide ( $\text{AlO}_x$ ) to form the Josephson junctions. The most



*A historical perspective of numerous improvements of the superconducting qubit lifetime over the past decade. All of these improvements are the result of circuit design instead of improvements in the constituent materials. (Devoret 2013)*

common method for forming the Josephson junction employs shadow evaporation of aluminum, and subsequent oxidation of the Al to form a thin, few-nanometer-thick insulating barrier, followed by a second evaporation of aluminum. The maximum tunneling current is determined by the effective barrier transparency, which is affected by the quality of the Al/AlO<sub>x</sub> interface as well as defect density in the aluminum oxide. Thus, the Josephson junction quality is a function of processing hygiene and material quality. Limited efforts have investigated Josephson junction fabrication using alternative superconducting metals; however, this work has not been sustained. Moreover, there has been some interest in developing alternative nonmetallic superconductors such as transition metal nitrides. Titanium nitride is interesting because it is already a material used in standard cleanroom processes.

Over the past 10 years, there have been significant improvements in the performance of superconducting qubit devices. Coherence times, more specifically the relaxation time,  $T_1$ , and dephasing time,  $T_2$ , have increased approximately 10,000-fold. Despite these impressive improvements,  $T_1$  and  $T_2$  for superconducting qubits are still 1–2 orders of magnitude below what is necessary for quantum computing. These improvements have been realized primarily by improving circuit design; not by realizing material improvements. For example, the 3-D transmon utilizes a 3-D microwave cavity to maintain a strong coupling as well as suppress qubit decoherence by providing a clean electromagnetic environment, but the increase of an order of magnitude in  $T_1$  was not realized by purposely fabricating higher quality Josephson junctions. It is therefore recognized that improvements in the materials in addition to engineering the environment will be critical<sup>8</sup>.

The current approach to improve fabrication processes of superconducting quantum computing circuits is Edisonian and slow. The rate of improvements in the state of the art for Josephson junction structures can be accelerated using an integrated approach of synthesis, characterization, and simulation to address fundamental materials issues. Currently, it is recognized that superconducting qubit performance is limited by extrinsic factors (e.g., processing contamination, unwitting oxidation during fabrication, etc.) and intrinsic defects (e.g., two-level-systems, flux noise, etc.). Thus, traceable methods for synthesis and characterization need to be developed to mitigate sources of decoherence. For example, the intrinsic quality factor could be improved by using epitaxial aluminum, rather than evaporated aluminum.

Development of realistic models and simulations in the quantum regime is needed to iteratively and progressively improve fabrication of superconducting qubits. This methodology could also be used to understand intrinsic sources of decoherence. Another avenue for further refinement is the poorly understood microscopic origin of dielectric loss through two-level-systems (TLS). Accurate modeling would be useful in understanding the microscopic details of TLS noise sources when combined with systematic synthesis studies and loss data. Such data could be obtained from resonators utilized as sensitive detectors of intrinsic sources of decoherence. This approach has already paid dividends in improving coherence times of superconducting qubits, and needs to be further leveraged in the pursuit of the next breakthroughs in coherence time improvements.

Qualitative advances in superconducting qubit performance are possible if extrinsic sources of disorder and resulting decoherence can be identified and ultimately

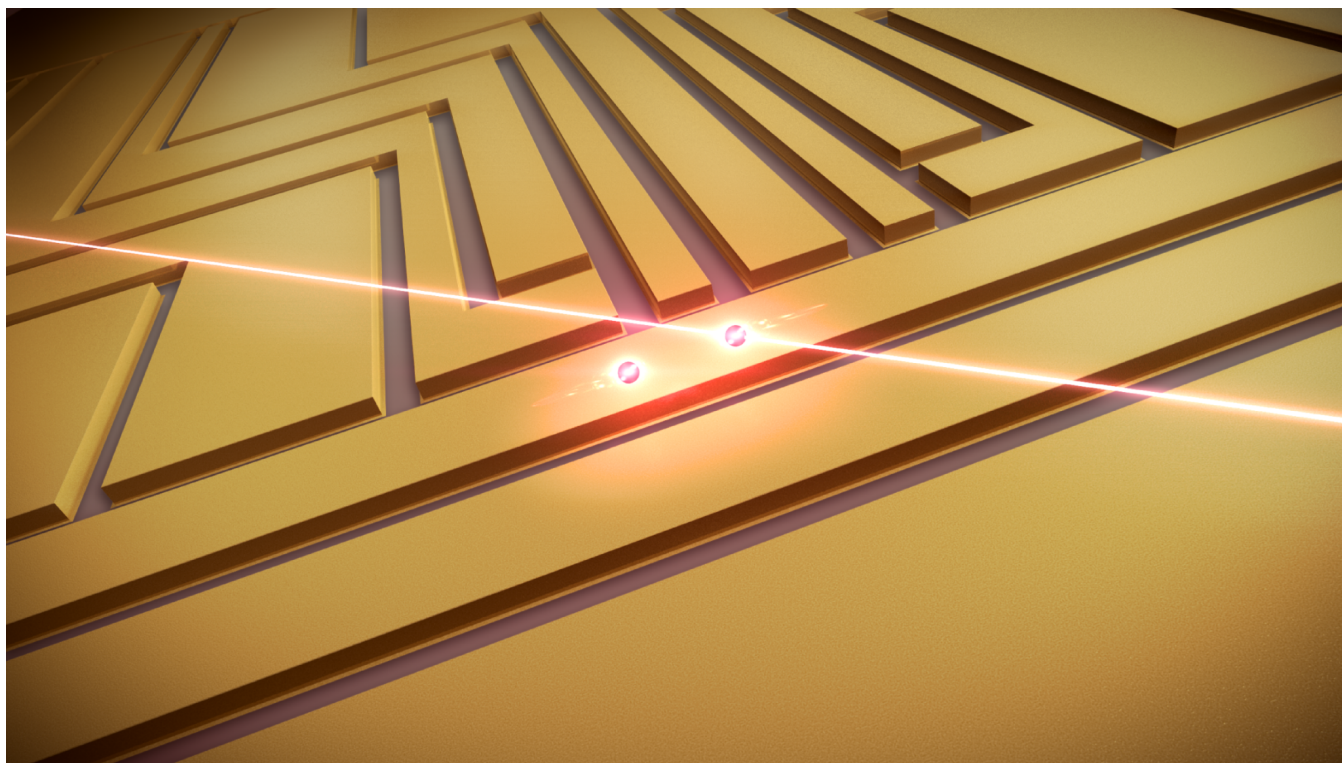


controlled. An integrated and systematic approach to synthesis, characterization, and modeling can relate measured defects to sources of decoherence by directly relating qubit functionality to process variables. It would also be useful to apply this integrated approach to identify alternative superconducting materials that meet the requirements for high-fidelity qubits, perform extensive analyses of insulating materials and tunnel barrier materials, determine if crystalline superconducting films are necessary to reduce intrinsic noise sources, and understand contamination and oxidation of interfaces during fabrication and substrate preparation.

### ***Ion Trap Qubits***

In the quantum computer architecture built around trapped ions, the qubit is an ionized atom which by itself is highly stable. The process of isolating and manipulating this ion

is so selective that the ions are always single atoms of known composition and ionization state. This is realized in an experimental apparatus that uses electrically conducting surfaces in close proximity to the ion and a laser beam that needs to be precisely controlled. Atomic ions, trapped in free space, can be initialized, entangled to perform quantum gates, and measured with nearly 100% efficiency. Trapped ions have outstanding coherence properties and form the basis of a compelling architecture for quantum computing. The atomic ions are confined in a UHV chamber in close proximity to trap electrodes by ponderomotive forces generated with a combination of radio frequency (RF) and direct current (DC) electric fields. The ions function as qubits after they are motionally cooled by ultraviolet laser light pressure into their ground state.<sup>9</sup> Because of the high accuracy of this approach, experimental demonstrations of interacting qubits are readily achieved.



*Illustration of a pair of ions cooled by a laser beam and trapped in an electrostatic trap created by the gold traces. (Hite 2014)*

Two problems associated with ion trap quantum computing are slow gate speeds and low spatial density of trapped ions. The use of smaller traps solves both problems, as smaller traps reduce the distance that ions need to move, thus increasing the rate at which ions can be coupled to perform 2-qubit logic gates. The smaller distance increases trap frequencies, thereby reducing gate times, and also allows for a higher ion density by enabling arrays of multi-node traps. Complexity of the laser driven fields necessitates improvements in optical materials and fiber optics, that allow for improved beam control. Additionally, efficient separation of ion qubits for transport between separate nodes requires smaller traps with good electrode surface integrity.

Smaller traps result in smaller distances between the ions and the electrode surface. However, disruptive heating of the ion from its motional (quantum) ground state can occur at distances of tens to hundreds of micrometers from a surface, which are also the requisite distances for the necessary gate speeds and ion densities. This motional heating of the ion from its ground state is due to fluctuating electric fields that are uncorrelated to the RF drive of the trap. This ion motional heating is referred to as “anomalous heating,” and the anomalous heating rates are much greater than what would be expected from Johnson noise or black-body radiation. Although the heating rates appear to follow a quartic inverse scaling law with respect to distance<sup>10</sup>, the fundamental heating mechanism is poorly understood despite extensive characterization<sup>11</sup>.

Although the underlying physical mechanisms for ion trap anomalous heating have not been explained, mitigation strategies are being investigated and employed. These include reducing the

anomalous heating rates by performing ion qubit coherent operations at cryogenic temperatures. Cryogenic operation has been shown to provide two orders of magnitude reduction of anomalous heating rate temperatures but confer a severe penalty in experimental complexity. Additionally, and more desirable experimentally, recent demonstrations have shown that in situ surface treatments of trap materials can help reduce anomalous heating rates. Ultimately, the optimal solution of anomalous ion heating awaits a better understanding of the cause. Fundamental materials studies and first-principles modeling of the ion heating should accelerate the understanding of the physical mechanisms of heating and help to find the best mitigation strategies.

The complexity of the laser-driven fields requires advances in optical materials and fiber optics to improve beam control. Materials challenges related to improving ion trap qubit technology are therefore focused on the electrode surface and improving the performance of the materials used to create and deliver the laser beams.

## **Opportunities for Materials Breakthroughs**

Improvement in any of the mainstream quantum computing technologies requires advances in materials science. Improvements in the performance and reliability of qubits require simultaneous and coupled advances to the state of the art in modeling, synthesis, and characterization. A comprehensive approach is needed to develop the capability to deterministically place atoms with minimal disorder, verify purity and validate structure. From the onset it seems daunting to control the placement of every atom in a complex qubit device, but by simultaneously connecting simulation with synthesis through characterization, the essential

material structure may be controlled to effectively remove seemingly random variance from the environment experienced by the qubit. By focusing efforts on the most critical atoms that are in the most critical locations, progress can be accelerated.

Decoherence and loss are primary concerns in each of these qubit technologies, as both limit performance by causing decay of the quantum state that is central to qubit functionality. Better understanding of how qubits interact with both deterministic and uncontrolled material influences on the quantum environment will allow such limiting mechanisms to be predictively understood and controlled. Such control will enable the quantum computing community to move beyond current mitigation strategies that simply attempt to decouple the qubit from its local environment. From the perspective of quantum computing, decoupling the qubit from its environment is counter-productive because the decoupled qubit is insensitive to the fundamental requirements of initialization, manipulation, and measurement.

Atomic- and meso-scale disorder in the atomic arrangement and interfaces between materials perturbs the local electronic properties and limits qubit performance. One form of disorder is unintentional extrinsic impurities that couple to the qubit. Current understanding of fundamental interactions between quantum states and the local environment is largely defined by our

knowledge of atomic-scale properties for the surrounding material. This understanding needs to be expanded to all scales in order to incorporate both extrinsic and intrinsic sources of disorder. Modeling is needed to relate sources of noise and disorder to impacts on performance so that fabrication and characterization advances can be strategically focused in the most efficient manner to accelerate device improvements.

Research in materials science can empower quantum computing by replacing empirical serendipity with predictive tailoring of materials properties in the quantum regime. Success will require materials modeling at all length, time, and energy scales with complementary and coupled advances in fabrication that decrease intrinsic and impurity disorder as well as discovery and development of next-generation characterization capabilities to resolve materials characteristics that are relevant to qubits and their environments. The synergistic coupling of characterization, synthesis, and modeling is essential for achieving advanced functionality in challenging quantum computing environments. While the challenges are manifestly hard, success will not only enable advances in quantum computing but also transformational breakthroughs with great applications at the frontiers of materials research.

## Grand Challenges/Science Questions

The present report describes significant quantum computing materials research opportunities and the most promising emerging directions identified by the workshop attendees. These research areas are described in the next section. Here we emphasize three grand challenges that cut across each of the directions and span multiple qubit technologies. These grand challenges are best addressed by collaborative research combining expertise in simulation, characterization and synthesis spanning traditional disciplines of materials science, chemistry, condensed matter physics, mathematics, and quantum physics. Because of their interdisciplinary nature, these challenges defy simple categorization within these traditional discipline boundaries.

### **Mechanisms That Drive Fundamental Physics Interactions Across Scales**

In order to develop rich models that preserve relevant physics across multiple scales of space, time and energy, our understanding of surface and interface physics must be refined to precisely prescribe the functionality of electronic structure on the micro-eV-scale and physical structure on the nanometer-scale. Our desire to quantitatively connect the microscopic interactions to device performance at the mesoscale is driven by recognized gaps in our understanding of how qubits interact with the local materials environment. This understanding must be extended to controlling the noise properties of isolated quantum states, as sources of noise are key roadblocks to quantum computing.

Noise in physical qubits limits the coherence of their quantum states and corrupts entanglement among qubits, reducing the system reliability. Many decoherence processes are intimately related to the materials and processes used in their fabrication. The materials-sensitivity of qubit performance has become very apparent. Uniform, controlled fabrication of high-quality, long coherence time qubits requires predictive understanding of the relationships between fabrication processes, microscopic materials properties, qubit performance, and multiple qubit interactions. In addition, predictive understanding of interactions between fabricated qubits is essential to evaluate spatiotemporal correlations and crosstalk of the decoherence processes, enabling design of optimal fault-tolerant systems.

An integrated and experimentally-validated modeling capability reaching across length and phenomenology scales will enable controlled fabrication of reliable, reproducible, high-fidelity qubit systems. Such a predictive modeling framework is required to advance the current state of the art beyond abstract, average noise models in gate-level simulations so that a direct connection to specific devices and their fabrication can be made. This missing capability is analogous to the well-established methodology in the CMOS industry of using highly predictive models of fabrication processes coupled to device-scale performance for the integrated design of microchips.

An integrated modeling approach should begin at the atomistic level with *ab initio* methods to predict materials- and fabrication-specific microscopic sources of noise and decoherence in real qubits, with



implicit dependence on fabrication and environmental variables. Further insight into fundamental microscopic origins of the qubit-material interactions and emergent interactions at larger scales are important in understanding the dynamics of a complex qubit technology. The atomistic models should feed parameterized mesoscale noise models that can ultimately be embedded in device-level models to prioritize sources of noise for characterization and control. Revealing intrinsic properties as distinct from noise resulting from technologically limited disorder signatures will enable device-level simulations that incorporate smaller scales to predict the response of qubits and allow simulation to advance a key technological frontier.

### **Engineered Materials With Exquisite Control of Electronic and Physical Structure**

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Designing next-generation materials for quantum computing requires more than substituting different atoms in familiar structures. Instead, researchers need to understand the limits of atomic order and then control disorder through precise manipulation of material structure. In addition to quantifying and predicting the role of processing disorder on performance, other issues such as understanding and optimizing the limitations of synthesis and extending the technological limitations of creating perfect structures must be addressed.

Creating novel materials is at the heart of materials science, and the desire to arrange atoms in a unique fashion that provides a new function or performance standard is strong in the research community. It needs to be recognized, that next-generation materials for quantum computing require both

creative, new, groundbreaking heterogeneous materials, and materials improvements that result in advancements of the order and purity of emerging materials.

Quantum computing device performance is also strongly influenced by design and fabrication processes. Advances in in situ characterization during synthesis and fabrication, combined with integrated and collaborative teaming among fabrication, characterization, and simulation researchers, will be essential to achieve the necessary control to produce materials that are beyond current CMOS-quality materials.

Mesoscale features resulting from intentional and inadvertent clean room processes is an area that offers the potential for dramatic acceleration of the state of the art. Microscale features fabricated with near-atomic precision are at the resolution limits of today's most advanced lithography tools. The challenge of fabricating devices with extreme critical dimensions may also be compounded with emerging challenges related to manipulating atoms one at a time in a deterministic manner and may employ both bottom-up and top-down fabrication approaches.

### **Techniques to Tease Out Novel Properties of Materials at Unprecedented Resolution**

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Transformational advances in measurement sciences comprise the third pillar towards achieving insight into the intrinsic phenomena of qubits interacting with the local material. In this context, the role of advanced measurement science is to guide materials modeling towards accurate descriptions of time- and length-scales of value to quantum computing device measurement. By nature, measurement of

qubit materials and devices is a grand challenge for the most advanced characterization tools available. Combined characterization and simulation are needed together to pioneer measurements that will discover degrees of freedom and control relevant to individual quantum states. With great advances in applying computational analysis to atomic-scale resolution techniques, measurement techniques must keep pace and be extended beyond current measurement limitations.

Novel approaches are also needed. Today's quantum computing technologies are perhaps the most direct and sensitive sensors in existence. Instead of focusing solely on

creating quantum computing devices, one promising approach for characterization involves harnessing isolated quantum states as measurement probes to characterize local material. Through a variety of modalities, insight may also be gained through measurements of dynamic phenomena that supplement our understanding of systems at or near equilibrium. Furthermore, advancing techniques that enable characterization of germane materials properties without fabricating entire qubit devices, and perhaps, without cryogenic testing offers a valuable means to accelerate an integrated approach of synthesis, characterization, and simulation.



## Transformational Opportunities/Thrust Areas

Motivated by the importance and challenge of quantum computing and inspired by the materials grand challenges that hold promise for meeting this need, workshop attendees identified the following priority research directions for advancing materials research and empowering quantum computing:

- Sensing the local quantum environment through “qubit spectroscopy”
- Predicting microscopic origins of qubit material interactions
- Controlling atomic order exquisitely
- Imaging qubit materials in real time and multiple dimensions
- Connecting atomic fundamentals to qubits through mesoscale modeling
- Revealing qubit dynamics and decoherence mechanisms
- Cultivating the precise fabrication of ultrapure qubit devices
- Authenticating qubit devices using integrated models
- Eliminating deleterious environmental effects through advanced materials manipulation

### Sensing the Local Quantum Environment Through “Qubit Spectroscopy”

Qubit devices are routinely used to baseline performance and gain insight into the effective environment of the qubit. This is an important, direct, and very sensitive means of evaluating loss and decoherence mechanisms in qubits, but it is also an inefficient method of evaluating materials properties and the impact of fabrication processes on performance. An opportunity exists to invert this approach, using qubits as

spectroscopic detectors of quantum materials functionality.

#### **Opportunity**

Reducing decoherence and loss rates are primary research goals in quantum computation. Advancement in these areas would constitute measurable progress towards improving qubit performance. To this end, different materials are explored and processing splits are performed, but in general, only examples of positive variants are reported, while trends and specific connections between various levels of performance and specific processing defects are not.

There is a clear opportunity to leverage functioning qubits at all levels of performance as sensors to quantitatively analyze device performance in relation to the quality of the constituent materials and the specific processes from which it is made. In order to leverage the sensitivity of qubits as sensors of the quantum environment, devices need to be designed that are tunable, or designed in such a fashion to probe the material instead of optimizing for ultimate performance of maximizing state lifetimes and coherence times. The correlation of these quantum-sensing measurements to more conventional materials characterization techniques will enable accelerated advances in relating materials processing to qubit performance.

#### **Timeliness**

Superconducting qubits are made from a variety of materials such as aluminum, niobium, titanium nitride, and even rhenium. Edisonian assessments of these different material systems are completed to determine the apparent impact of process variations<sup>12</sup>

and material variations<sup>13</sup>. In superconducting qubits, dielectric loss mechanisms have been described by the TLS loss model that is an abstraction of these materials as a pair of harmonic potentials<sup>14</sup>. Another loss mechanism that has been identified is 1/frequency flux and charge noise that have universal scaling characteristics<sup>15</sup> and dependence on processing history.

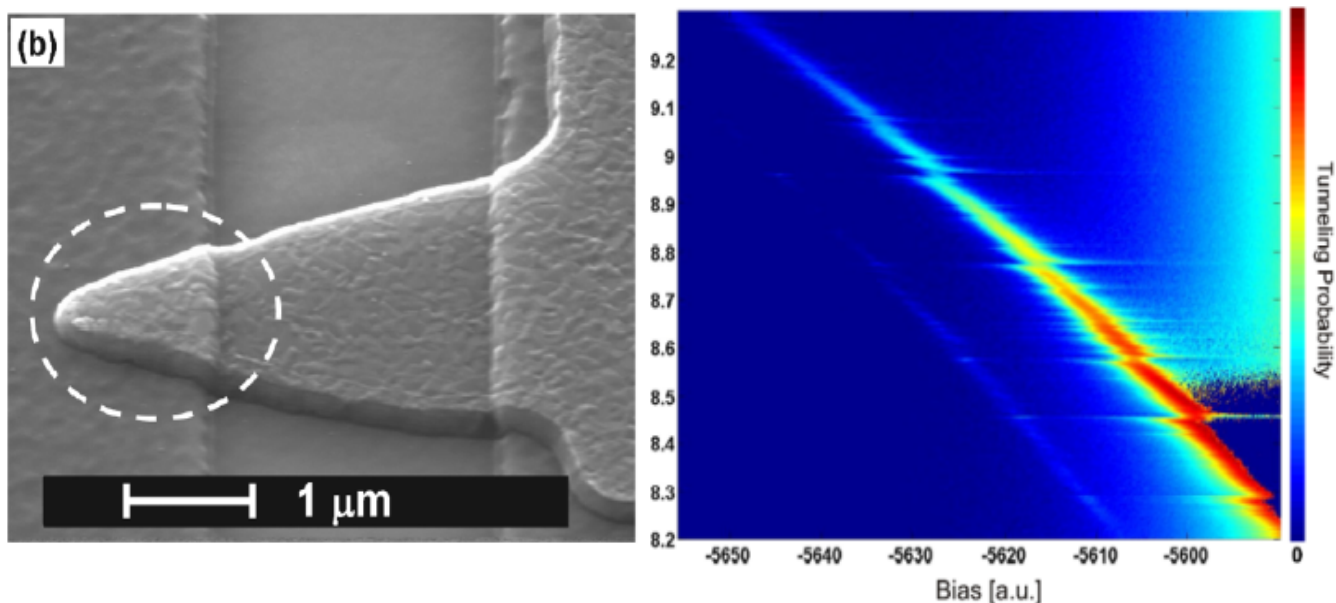
Viable semiconductor qubits are all fabricated from silicon and silicon germanium alloys to enable fabrication from material that can potentially be free from nuclear spins. Varying the sensitivity of the qubit to charge noise as a function of qubit structure and gating strategy could potentially be used, not to isolate the qubit from the environment, but rather to energetically probe the local device. Through appropriately designed devices that accentuate key characteristics, qubits can be used to quantify aspects of the device and also to extract the charge noise spectral density of the device, density of noise fluctuators at various interfaces, and/or probe the local band potentials to enhance understanding of valley potentials.

With ion trap quantum computing, the only currently used measure of anomalous heating is with the ion itself. Current ion trap qubit spectroscopy experiments explore

correlations between materials that are cleaned via different techniques, different surfaces, and temperature dependence of this phenomenon. The goal of such experiments is discovery of the phenomenon in order to discover the underlying physical mechanisms underpinning anomalous heating of ions that are in close proximity to a solid surface.

### **Approach**

One difference between designing a quantum sensor and a qubit is the intent. Sensor applications explore performance as a function of a tunable parameter, while a qubit maximizes performance with respect to targeted parameters. Currently, tuning is typically implemented by changing the excitation energy, flux bias, frequency, and/or temperature. For quantum sensors, the concept of a tuning parameter needs to be broadened. Whether it is the coupling coefficients, capacitive charging energy, Josephson coupling energy, participation ratio, or degree of valley splitting—experiments need to be designed that test the trends of an isolated qubit interacting with the surrounding material in a deterministic fashion. Furthermore, for all quantum technologies under consideration, qubits can be realized experimentally fairly commonly, but not necessarily reproducibly. This variation in itself can be used as a probe.



*Electron microscope image of a sub-micron Josephson junction fabricated with shadow evaporation. The measured “noise” in the tunneling probability results from undesirable two-level systems that are energetically similar to the qubit energy. Small junction or improved materials are needed to reduce these features. (Steffen 2006, Pappas 2014)*

Many classical materials characterization techniques may find quantum analogues through quantum sensors. For instance, near-resonant pump-probe spectroscopy is routinely performed by the quantum computing community to measure dynamic qubit responses. It is possible that in addition to these resonant techniques, non-resonant dynamic probes may provide insight into the energy landscape of quantum computing devices. Teaming scientists with expertise in materials and quantum computing is needed to leverage and bridge the capabilities and experience from both disciplines in order to fully develop and exploit this concept of qubit spectroscopy for the optimization of qubit materials.

Success in this approach will also require an understanding of why a qubit performs poorly as opposed to optimizing qubit properties. The focus on “bad” qubits and not just “good” qubits as variable probes of matter is in stark contrast to the usual emphasis in quantum technologies. However,

characterization and study of “bad” qubits will accelerate our understanding of the relationship between materials parameters and qubit performance. This approach will provide much needed input to models describing qubit performance as a function of material structure and qubit device design.

### **Potential Impact**

Expanding the use of quantum devices as sensors of the local material system is the most direct means of evaluating materials in the qubit operating regime. Quantum sensors have already been demonstrated as extremely sensitive probes of other physical parameters, for example gravity sensors based on trapped atoms. Developing quantum devices as materials sensors will directly impact the quantum computing community, improve the understanding of defect phenomena in materials, as well as advance the frontiers of materials prediction and control.

## Predicting Microscopic Origins of Qubit-material Interactions

Simulations based on fundamental theory can predict atomistic details of materials that affect the sensitive performance of quantum devices. Predicting the origin of the fundamental interactions between a qubit and the material can lead to enhanced control of materials and refined device fabrication processes.

### **Opportunity**

Specific details of materials-related noise characteristics and device-level interactions depend on the qubit technology. Various known and unknown microscopic mechanisms exist that connect the atomistic structure of materials and their imperfections to noise and decoherence processes. However, the origins of noise are largely undiscovered in the multitude of atomic structures that comprise real materials. These include the atomistic structure of surfaces and interfaces, defects and impurities in the bulk and at interfaces, as well as the intrinsic electromagnetic properties of materials and surfaces. The dependence of noise characteristics on fabrication conditions and post-fabrication exposure of devices is explicit and must be identified and characterized.

The transition from ideal-to-real materials modeling at the atomic scale requires the ability to explicitly account for defects, impurities, non-perfect interfaces, microstructure, and environmental effects in a large phase space with large numbers of atoms. For example, in superconducting quantum computing, the TLS noise sources that matter most are those with excitations in the same energy range as the energy splitting of the qubit with which they couple, which is on the order of  $10^{-5}$  eV. Such a fine energy level structure challenges the current

capabilities of ab initio or classical direct computer simulation of electronic and vibrational excitations. For example, if the density and characteristics of TLS noise sources at a given metal–oxide interface could be predicted realistically, then computationally designed interfaces could be explored *before* fabricating them. New modeling approaches are clearly necessary to achieve such a goal.

### **Timeliness**

Recent progress in first principles theories of condensed and molecular systems has opened up the possibility of using highly accurate quantum materials simulation methods to address the complexity of materials at a detailed, microscopic level. Such methods may consist of density functional theory (including advances in exchange-correlation functionals), quantum Monte Carlo, GW perturbation theory, and quantum chemistry techniques. Petascale-class and beyond supercomputers often need to be employed, along with existing and under-development codes that can take advantage of such highly parallel computing platforms. In addition, recent advances in the fabrication of high-quality individual qubits enable direct connection and iteration with experiments to identify and control the atomic-scale features of materials that impact qubit operation, as well as provide validation for advances in computational methods toward higher accuracy.

### **Approach**

Atomistic simulations using ab initio methods can be used to predict the materials- and fabrication-specific microscopic sources of noise and decoherence in real qubits, including the implicit dependence on fabrication or environmental variables. The sources of noise may comprise various external spin, magnetic, or tunneling TLS

noise sources that can couple to the qubit and are manifested as crystal defects, bulk or surface impurities, and even imperfect bonds in amorphous structures. Using large numbers of small-to-medium-scale simulations that are much more efficient than conducting experimental tests holds promise to greatly accelerate our understanding. Combined with a smaller numbers of large-scale simulations, the fundamental parameters needed for mesoscale computing and device modeling may be determined.

Future advances in ab initio atomistic simulation methods need to be leveraged and/or developed. Density functional theory is the workhorse for such simulations, but more advanced methods improve the accuracy of predictions that are needed for simulating the effects of value to quantum computing. Progress is required in the state of the art to reach larger length scales and relevant energy scales approaching micro-eV are required. Highly scalable and reduced scaling approaches that can take advantage of millions of cores on modern supercomputers to address system sizes approaching 10-100K atoms are desired, as are computationally efficient electronic structure approaches—including next-generation quantum Monte Carlo and GW techniques—for going beyond density functional theory routinely. Advances in sampling algorithms and techniques to bridge multiple length and time scales may be leveraged both for microscopic and mesoscale simulations.

The simulation framework should be integrated as tightly as possible with experiments. The integrated models must provide a direct connection between the details of fabrication processes, defect distributions, predicted materials properties, and device response. The models can then be used to design and interpret specific

experiments on real devices to allow model validation and ultimately determine the dominant noise mechanisms. Experimental probes of material compositions and atomic structures enable comparison between experiments and models through specification of the material parameters. Iteration of modeling and experimentation should lead to optimized device and circuit fabrications.

### ***Potential Impact***

The current missing link between atomic-scale models of materials and device-level models of noise hinders the rational bottom-up design of robust, reliable systems of qubits. The ability to use predictive materials simulations on petascale and beyond computing platforms represents an opportunity to accelerate the discovery, development, and optimization of new materials for quantum computing technologies. Coupling these advanced materials simulations to device models will allow direct prediction of device performance and correlate gate error rates with fabrication processing variables. Optimization and control of the fabrication process to achieve optimal system performance is a key objective that is enabled through simulation-based research in the microscopic origins of qubit-material interactions.

Advances such as these will result in powerful improvements and refinements in the computational “toolbox” that will be applicable to various materials systems. Enhancements in energy resolution, increases in simulation size and timescale, and extensions of scaling approaches will yield greater insight into more complex materials behavior that will impact simulations of electronic materials across a broad application space.

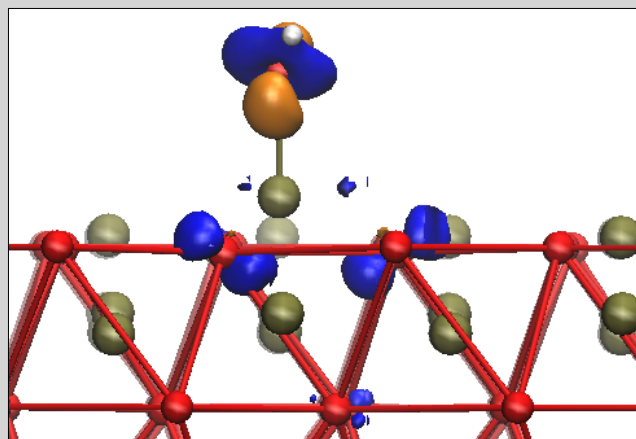
## Identifying Atomistic Sources of Decoherence in Superconducting Qubits

While a number of theories have been proposed to explain the mechanisms of quantum state decoherence in qubits related to materials defects, the identification of specific defects that are responsible in particular materials is required for rational process variations to be devised to reduce or eliminate them and improve qubit performance. Many of the proposed mechanisms focus on defects in oxide insulator materials and often involve effects at surfaces or interfaces. Recent works have used atomistic simulations to identify some candidate impurity-related defects in aluminum-oxide materials (sapphire and alumina) that can introduce magnetic or microwave resonant noise in superconducting qubits (SQs). Aluminum oxide is commonly found in many state-of-the-art SQs, as the sapphire substrate material, the Josephson junction insulator material, or the native oxide on aluminum superconducting lines.

Various models appear in the literature that implicate surface or interface spins for the decoherence of SQs, including a model that conjectures hopping of electrons among trap sites with different preferred spin orientations, the involvement of potential fluctuations at metal-insulator junctions, or paramagnetic dangling bonds at various materials interfaces that can flip their spin by interacting with nearby lattice excitations (phonons). In all these models, the specific responsible microscopic object for a given material is unknown or unspecified. Impurities on the surface may also be paramagnetic and introduce magnetic flux noise.

A recent paper by Lee, et al.,<sup>16</sup> investigates the possibility of impurities from the environment adsorbing onto sapphire or alumina and creating local paramagnetic defects. Using density functional theory, the authors find that

water vapor can readily dissociate onto alumina and under certain conditions create an isolated adsorbed OH defect with a fluctuating magnetic moment (Figure 1). These defects can cause significant noise in flux qubits fabricated even with single crystal substrates, but can also couple to aluminum superconducting lines in resonant devices from the native oxide that grows on the metal surface when unpassivated. In addition, differing conditions of humidity or heat treatment during fabrication can cause variations in the observed device response. The researchers discovered that such detrimental OH impurities on alumina may be displaced by less electrophilic chemical groups that do not exhibit the paramagnetic behavior;  $\text{NH}_2$  was suggested as a possible choice that binds to the surface more strongly than OH. Further work is required to experimentally validate the models and also explore additional materials and impurities.

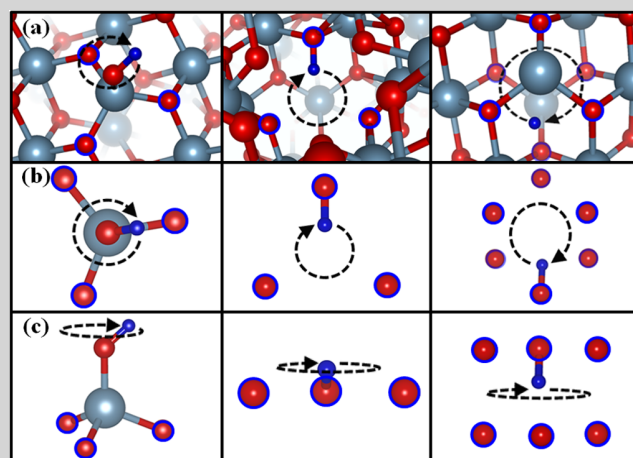


**Figure 1.** The electron spin density (blue and orange isosurfaces) associated with a paramagnetic hydroxyl impurity defect on the sapphire surface. (Taken from Ref. 1.)



In resonant cavity SQs, tunneling two-level systems (TTLS) associated with structural variations in amorphous materials have been conjectured as a source of noise. The precise mechanism is still not fully understood, but involves resonant coupling of the microwave cavity photons with the TTLS defects. Increasingly sophisticated models have been proposed, involving possible RKKY interactions among dilute spins at an interface; interacting two-level systems with a spectrum of fluctuating resonant energies arising from direct coupling, RKKY interaction, elastic strain, or other unidentified interactions between spins; or fluctuations associated with metastable magnetic clusters of both ferromagnetic and paramagnetic character, driven by interactions between TTLS from potential gradients at a superconductor–insulator interface. As above, the materials-specific identity of such objects is not known. Recent works by Holder, et al. and Gordon, et al.<sup>17</sup> using density functional theory have shown that H-related defects in crystalline or amorphous materials may contribute. By computing the potential energy landscapes of H rotor defects in alumina and solving for the quantum mechanical tunneling rates (Figure 2), Holder et al. found frequencies that span the GHz range typical of qubit resonant cavities for possible defects consisting of OH surface rotors, hydrogenated Al vacancies, or H interstitials. The calculated dipole moments of the defects were also consistent with experimental determinations. However, the accuracy of the frequency calculations was not sufficient to be able to distinguish the different defects. Gordon, et al. also used density functional theory to analyze hydrogen interstitials in  $\text{Al}_2\text{O}_3$  and similarly found that when in a hydrogen bonding configuration between two host oxygen atoms, resonant tunneling frequencies are in a range that may couple to qubits. The effect was found to depend on the O–O distance in the host, which

correspond to typical distances in amorphous alumina or in near-surface regions of crystalline  $\text{Al}_2\text{O}_3$ . As above, opportunities exist to refine such calculations and also expand the exploration to a wider set of defects and materials.



**Figure 2.** Bulk and surface H rotor defects in  $\alpha$ -alumina considered as resonant tunneling two-level systems and investigated with density functional theory to predict their tunneling frequencies and local dipole moments. (Taken from Ref. 2.)

## Controlling Atomic Order Exquisitely

Many limitations of qubit performance can be traced back to disorder and impurities in the constituent materials from which they are made. The intrinsically quantum nature of qubit performance sets a uniquely high bar for increasing the refinement of materials and their means of production, driving the need for greater synthesis fidelity.

### **Opportunity**

Whether the synthesis tool is a state-of-the-art molecular beam epitaxy system, or a modern day chemical vapor deposition system, the creation of imperfect materials with disorder resulting from contamination or intrinsic defects has been linked to degraded qubit performance across all quantum computing technologies. Stringent requirements for bulk materials as passive substrates, interfaces in close proximity to the qubit, and thin films grown epitaxially or with an amorphous structure, are driven by the high sensitivity of qubits to the local environment. Ideally, interfaces that are truly atomically smooth and layers that have zero defect states may be needed to eliminate technical limitations of qubit loss and decoherence mechanisms. For both highly ordered and disordered materials microscopic *ab initio* simulations can provide critical guidance to accelerate process development, in contrast to the current Edisonian, trial and error, approach.

### **Timeliness**

The impact and limitation of high-quality materials are apparent in both superconducting and semiconductor-based quantum computing technologies. In superconducting quantum computing, epitaxial material, as compared to other microstructures, has shown significant improvements in reducing dielectric loss. It is

not clear if the microstructure itself impacts performance, or if the enhanced purity of epitaxial material is the root cause of these performance improvements. Only a few groups have mastered the technical difficulties associated with producing material that achieve loss tangents better than  $10^{-6}$ , and as a result, many groups are pursuing 3-D superconducting cavities, such as those developed for particle accelerators, because the 3-D cavity minimizes the role of material loss on qubit performance. The development of processes to yield ultrapure and extremely low-loss materials on low-loss substrates is a ripe area of research. Surprisingly, the incorporation of epitaxial Josephson junctions in superconducting qubit circuits has yet to result in substantial improvement of device performance, as has been observed for giant magnetoresistance applications.

For semiconductor-based quantum computing, the role of disorder is of key importance. Substrate miscut, impurities, and intrinsic growth defects are some issues that complicate the engineered valley physics needed to break the conduction band degeneracy. Additionally, low temperature overgrowth of silicon and germanium that can be implemented *in situ* with scanning tunneling lithographic microscopy could greatly enhance donor qubit performance.

### **Approach**

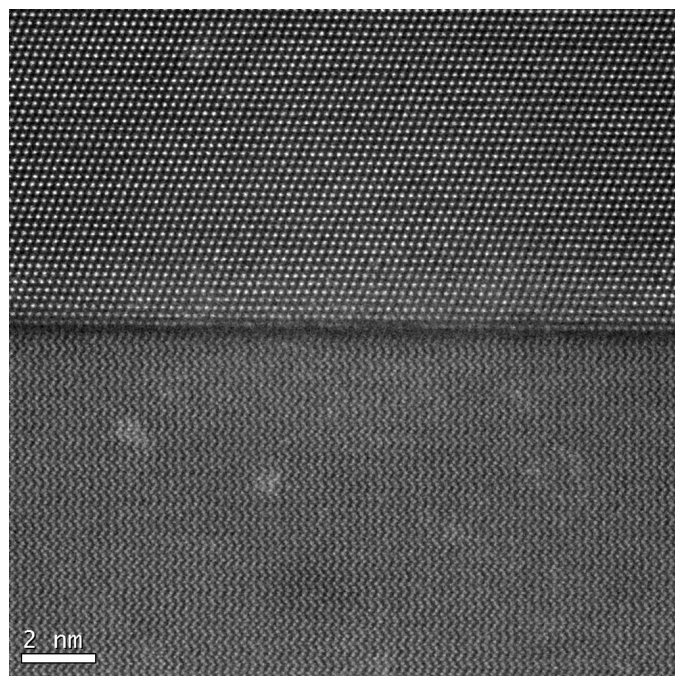
The ultimate performance of any engineered structure is limited by the quality of the initial material. Epitaxial materials grown with high purity techniques such as molecular beam epitaxy and chemical vapor deposition that have become established through the electronic and optoelectronic industries have been used to impact both superconducting and semiconducting quantum computing. Development of solid sources that can handle the high heat load of

low vapor materials, high purity gas sources and the means to safely handle them are examples of component improvements that may dramatically impact material quality. Expanding the use of these materials and using cluster tools to create multilayered structures without exposing samples to atmospheric environment are important steps that need to be adopted by the quantum computing community.

In order to produce next-generation materials, it is expected that technical advances in the equipment that is used to produce these materials will be required. Areas of importance include in situ diagnostics, vacuum connected systems or means of transferring samples between process tools, system design improvements to processing and growth tools currently used to fabricate high-quality qubit materials, and improvements in characterization tools that need to extend sensitivity floors beyond the current state of the art.

The pursuit of new growth technologies, sources, and materials needs to be carefully considered, as creation of a single crystal is not in itself sufficient. The fabricated materials must either evolve past the current high-quality materials, or provide a means that can disrupt current materials quality, providing an achievable pathway towards ultrapure, low disorder materials that outperform current materials.

Understanding the specific microscopic loss mechanisms and their structure-function relationship is key to guiding the discovery of new materials with improved purity. Research in growth techniques and strategic simulations must be accompanied by research in the most suitable characterization techniques. For example, semiconductor quantum computing commonly uses the low-temperature mobility of a two-dimensional electron gas



*Atomic resolution scanning transmission electron microscopy image of aluminum single crystal grown metamorphically on a sapphire wafer with an abrupt and perfect crystal interface. This image is a result of a spontaneous collaboration following the 2014 Materials Science Workshop at LANL. (Arslan 2015)*

material as a metric for the quality of the material. However, it is often the case that the best high-mobility material does not yield the best qubit performance. Other means of assessing disorder are required that are more applicable to the operating regime of the qubit, where few particles are present and the screening fields that exist in ensemble measurements do not participate. Development of such techniques will require a more detailed understanding of the interaction of the qubit with the local environment that is mediated by the surrounding material.

### **Potential Impact**

The ability to synthesize device-quality materials is a current bottleneck in quantum

computing. The development of processes enabling the fabrication of quantum grade materials would substantially accelerate progress in this arena. More widely available and more predictable qubit materials will bring more researchers to the quantum computing community. Next-generation fabrication tools coupled with advanced characterization techniques will enable advancement in this arena. This will accelerate not only quantum computing

technology specifically, but also materials by design in general.

While the specific structures needed for quantum computing devices are unique, the tools that are used to make them are not. These techniques developed to exquisitely control materials will enable enhanced performance of other functional materials for applications in conventional information technologies, sensors, energy storage, transmission, and transduction.

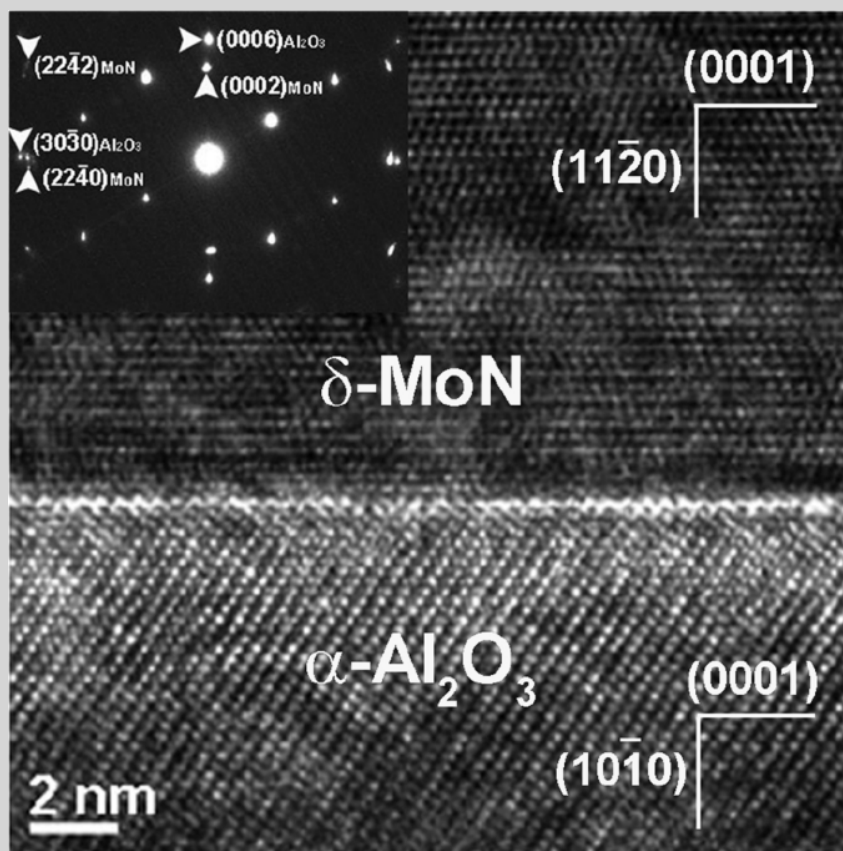
## Novel Epitaxial Materials and Synthesis

From a materials science approach, the most direct means of addressing the limitations of superconducting qubits related to decoherence is to design and grow new materials that offer reduction of the sources of noise. While there is much discussion regarding the specific microscopic origins of two-level system noise, it is generally accepted that highly ordered epitaxial materials offer great potential for realizing low noise materials. One such example is epitaxial aluminum on sapphire and silicon that shows marked reduction in the microwave loss-tangent compared to polycrystalline and amorphous aluminum on similar substrates.

Superconducting transition metal nitrides such as TiN and NbN are of current interest to the superconducting qubit community, and with this interest arises potential for alternative synthesis techniques that offer a means of circumventing the stringent system requirements associated with high melting temperature and low-vapor pressure materials such as Ti and the refractory metals, in order to produce materials with microwave loss-tangent values lower than  $10^{-6}$ , which is currently realized with both Al and TiN superconducting materials.

While molecular beam epitaxy, chemical vapor deposition, and sputter deposition are expected to play key roles in materials synthesis in the future, a promising potential path forward is demonstrated through the synthesis of superconducting crystalline  $\delta$ -MoN films grown on crystalline  $\text{Al}_2\text{O}_3$  using a wet chemical solution method. While the structure of these materials has been shown to be exceedingly good as determined by x-ray diffraction and cross-sectional high-resolution transmission electron microscopy, that path towards acceptance of novel materials is challenging. Emerging materials need to demonstrate a host of characteristics such as structural and chemical stability through conventional clean room processes, and superconducting circuit elements with low dielectric loss tangents in the vicinity of 5 GHz before adoption is likely.

Emerging synthesis techniques, combinations of films and substrates with care of interface chemistry and structure, are sophisticated problems that offer great potential benefit as possible disrupting technologies to all quantum computing devices.



Y. Zhang, N. Haberkorn, F. Ronning, H. Wang, N. A. Mara, M. Zhuo, L. Chen, J. Hwan Lee, K. J. Blackmore, E. Bauer, A. K. Burrell, T. M. McCleskey, M. E. Hawley, R. K. Schulze, L. Civale, T. Tajima, and Q. Jia, "Epitaxial Superconducting  $\delta$ -MoN Films Grown by a Chemical Solution Method," *J. Amer. Chem. Soc.* **133**, 20735 (2011).

*High-resolution transmission electron microscopy image of the interface between a  $\delta$ -MoN film and an  $\text{Al}_2\text{O}_3$  substrate, and corresponding selected area electron diffraction pattern (inset).*

## Imaging Qubit Materials in Real Time and Multiple Dimensions

Precise spatial and temporal characterization of material chemistry and structure are critical to understanding the electronic structure and functionality of quantum-grade materials. Atomic-scale characterization of dilute impurities and disorder in the bulk and at interfaces need to be resolved in order to connect uncertainties in the materials structure with qubit performance.

### *Opportunity*

The optimization of qubit architectures requires an agile and flexible suite of microscopies with different contrast modalities. In addition, combining destructive cross-sectional characterization techniques with nondestructive characterization methods will provide a more complete multidimensional and multi-modal picture of the qubit to facilitate structure-function correlation within a specific architecture.

An important first step is use of the currently best available methods and approaches to characterize existing qubits. A number of these next-generation materials characterization techniques are currently housed at many of the DOE national user facilities. These instruments are capable of not only imaging the complex qubit structures at atomic to nanometer resolution but also of providing rich chemical information and physical information on a variety of time and energy scales.

It is important to note that the data provided by these advanced characterization methods may dramatically improve our ability to model the materials that comprise specific qubit architectures. Measurements of the critical physical and chemical features of qubit materials and correlation of those

measurements back to its observed quantum behavior will help close the make-measure-model loop and ultimately lead to better, more robust, and scalable models. Using these techniques on a problem as challenging as qubit performance will also stretch the limits of their performance leading to further technique enhancements.

### *Timeliness*

The ability to move beyond static structural/chemistry characterization to study ultrafast, non-reversible, chemical and magnetic properties of novel materials with nanometer spatial resolution is emerging and represents a significant new opportunity with implications for qubit characterization. Indeed, key characterization goals for qubits cannot all be achieved using currently available high-resolution tools and sample-preparation requirements. Complications resulting from finite penetration depth of electron microscopes, spatial resolution limits with proton and neutron radiography, insufficient temporal resolution from proton, neutron, and electron techniques, the lack of chemical or magnetic contrast and resolution limiting x-ray optics in full-field and scanning x-ray microscopy all fundamentally limit the adaptation of these techniques to time and spatially resolved characterization of qubit materials and devices. Additionally, near-surface scanning probe techniques such as atomic force microscopy and scanning tunneling microscopy generally reveal limited or no information about bulk properties or buried interfaces. Increasingly, it appears that ultrafast, coherent x-ray sources provide an additional tool for dynamic probing of buried interfaces with nanometer resolution at femtosecond time scales<sup>18,19</sup>. This new capability now exists at advanced DOE photon-based user facilities, as well as on novel laboratory-based, tabletop sources.



Materials challenges such as surface roughness, chemical homogeneity at interfaces, chemical purity, surface contamination, dopant location and concentration within a structure, and lattice strain in multilayer structures arise in at least one if not several of the most attractive qubit technologies. All of these issues have direct impact on the quantum behavior of the material and its function as a qubit. As the quantum computing community strives to improve materials and devices, now is the perfect time to examine both high performing qubit materials and those that failed to perform as expected, with the goal of understanding the relationship between structure and performance.

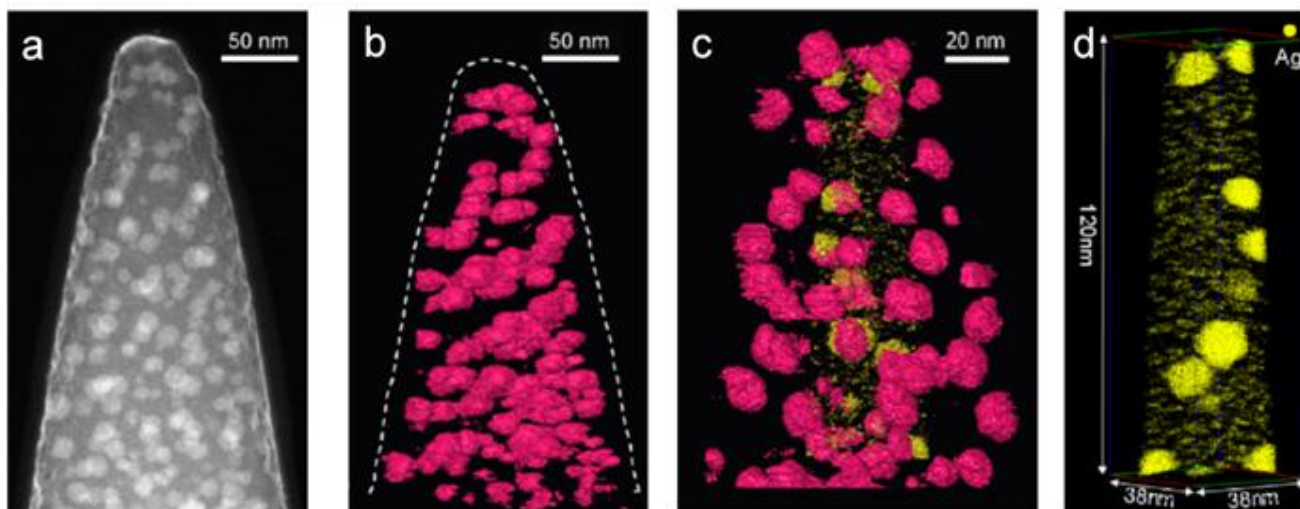
### **Approach**

Materials structures that form qubits are both small and complex in nature. In order to interrogate the fundamental physics of various qubit materials and structures, very high-resolution methods that can resolve individual atoms and atomic-level defect features must be used. One of the best methods for high-resolution analysis is scanning transmission electron microscopy (STEM). However, it is not sufficient to obtain only 2-D images or chemical spectra in 2-D; rather, high-resolution 2-D data must be complemented with 3-D imaging and spectroscopy to fully interrogate the qubit material. Accordingly, atomic-resolution STEM-based 3-D electron tomography, combining both atomic-level imaging and spectroscopy and mass spectrometry in the form of atom probe tomography (APT) can reveal essential complementary information.

When combined, STEM and APT can provide the full 3-D, near-atomic-resolution structural and chemical nature of the qubit, revealing insight on the localized disorder and defect structure of qubit devices.

While the scanning electron microscope (SEM) remains a powerful tool for metrology, there is little opportunity for further enhancements in its direct imaging performance. In contrast, the scanning helium-ion microscope (HeIM) is capable of 0.4-nm resolution under optimized conditions, offers a depth of field that is superior to that achievable by SEM, and provides strong topographic contrast. The advantages of the HeIM derive from the extremely short wavelength of a 30-keV He<sup>+</sup> ion compared to an electron<sup>20</sup>. Materials research in quantum computing may provide the “killer application” that requires the unique capabilities of the HeIM in order to succeed.

Scanning probe microscopy (SPM) has revolutionized nanoscale science and offers insight into the quantum nature of materials at an atomic scale. Scanning probe microscopies can broadly be divided into two categories based on the tip-to-surface interaction. In scanning tunneling microscopy (STM), feedback is provided by a tunneling current. In atomic force microscopy (AFM) and its many variants, surface imaging relies on force feedback.



*Correlation of STEM and APT reconstructions. a) STEM image of Al-Ag alloy. b) STEM tomogram of the alloy. c) Overlay of 3-D STEM reconstruction with APT reconstruction. d) APT reconstruction. (Arslan 2014)*

The STM is long recognized as a leading tool for surface structural studies and atomic manipulation, but perhaps its greatest strength is its ability to map electronic density of states near the Fermi energy with atomic resolution. Local electron density mapping is critical to identifying the quality and function of quantum structures that are used to define qubits. Further refinements to STM could include spin detection that may enable an understanding of the microscopic origins of noise that are predicted through simulation. The ability to probe and manipulate lattice, electron, and spin degrees of freedom with atomic-resolution would greatly enhance the understanding of qubit functionality.

The force feedback used in AFM makes possible the imaging of a number of local properties, including topography, conductivity, dopant levels, work function, Seebeck coefficients, and ionic transport that constitute the physical properties most often modeled in theories of qubit performance. Both STM and AFM have been adapted for measurements at millikelvin temperatures

and in ultrahigh vacuum. Cross-sectional techniques have also been demonstrated, which reveal atomic-level mixing at epitaxial interfaces, for example.

X-ray microscopy is still in its infancy but there are a number of attractive methods that can provide key information about local structure and composition, enabling the ability to characterize subsurface-materials that are close enough to interact with the qubit state. The use of advanced synchrotron sources, x-ray free-electron laser facilities, and tabletop sources for spectroscopy, scattering, and imaging is attractive because chemically specific absorption edges allow for deterministic chemical mapping. For the highest spatial resolution, scanning x-ray coherent diffraction imaging (XCDI), has demonstrated chemical sensitivity with high resolution for surfaces and interfaces. By means of iterative phase retrieval algorithms this technique can achieve a few nanometers resolution at large light source facilities or near wavelength-limited resolution (22 nm) from tabletop sources with much longer wavelength. This approach can potentially be

used for magnetic ptychography that promises subwavelength ( $< 1$  nm) resolution in structural and magnetic imaging. Furthermore, recent advances in tabletop systems may enable lab-scale experiments with unprecedented resolution, opening up applications with deeper penetration and chemical sensitivity than what is commonly available today.

Neutron characterization techniques (including polarized neutron reflectometry) offer unique magnetic and structural contrast that permits characterization of buried interfaces. Combining x-ray and polarized neutron reflectivity techniques offers insight into chemical and magnetic depth profiles that may lead to more advanced models and realization of quantum-grade materials.

Existing large-scale scientific facilities and infrastructure are poised to accelerate discovery and assist building new fundamental understanding of qubit behavior. While these measurement capabilities are not unique to studying qubits, the infrastructure that enables rapid sample-throughput and trains skilled instrument scientists also provides a fertile environment for initial exploration of the value of particular measurements to enhancing qubit functionality. These resources include synchrotron x-ray and neutron sources, scientific computing facilities, and nanoscale science research centers with electron-beam micro-characterization capabilities. The techniques available at such facilities enable the characterization of properties over a

range of length and time scales using a variety of modalities. Combining specular and polarized reflectivity techniques, for example, offers added insight into chemical and magnetic depth profiles that complement structural and electronic property measurements. The collaborative environments at these existing facilities are ideally suited to enable the validation of underpinning theoretical approaches.

### ***Potential Impact***

Multidimensional imaging of qubit materials across length, time, and energy scales must be pursued in all modalities. Techniques exist that probe materials at buried interfaces in order to study chemical composition, density, morphology, strain, and even electronic configuration. The scope and potential impact of all of these forefront capabilities will be expanded through their application to the understanding and optimization of materials underpinning quantum computation technologies. Furthermore, the unique capabilities contained within the Department of Energy national laboratories offer a completely new perspective for understanding the structure-function relationship of quantum computing materials with great potential for new science with a broad impact for understanding functional materials and their applications.

## Single Impurity Atom Imaging with Electron Microscopy

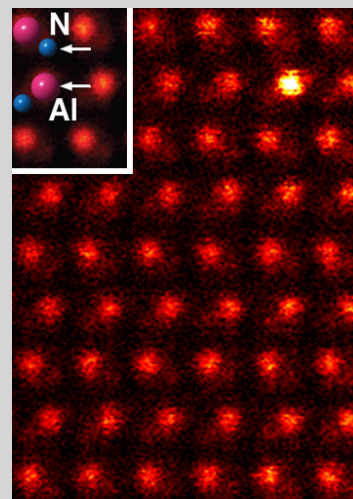
Designing systems that use a nano- or microscopic quantum device as the fundamental unit of information means that the surrounding environment needs to be understood and controlled with precision better than that of the characteristic states that are used to store information. Isolated defects and environmental defects that disrupt entanglement can dramatically affect qubit performance. Using current state-of-the-art characterization techniques to examine materials structure and local atomic motion, an important perspective is gained that provides opportunities for experimental verification of computational materials science simulations at the nanoscale.

High-resolution electron microscopy is one of the few techniques that have the resolution and sensitivity to detect a single impurity atom in an ordered material. Aberration-corrected imaging electron microscopes can achieve sub-Ångström resolution with a narrow depth of field that is on the nanometer scale. Combined with state-of-the-art sample preparation techniques, this capability enables imaging of impurities and defects in the bulk or at an interface.

High-angle annular dark-field (HAADF) imaging is an imaging mode that exhibits accurate Z-contrast images, where Z is the number of protons found in the nucleus, and therefore identical to the charge number of the nucleus. This sensitivity to the nuclear charge and high spatial resolution enables the location of a single dopant atom to be quantitatively determined. Examine the HAADF image of a wurtzite-aluminum nitride crystal as viewed along the  $[11\bar{2}0]$  zone axis. The atomic dumbbells consist of bright aluminum columns and less bright (almost invisible) nitrogen columns. In the upper right hand corner of the

image one column is brighter than the rest, which corresponds with a single cerium atom that has a higher nuclear charge than either aluminum or nitrogen. From this image it is apparent that this Ce atom was incorporated into an otherwise perfect AlN crystal by substituting for an aluminum atom.

*The HAADF image of a single Ce substitutional impurity atom (bright spot in upper right corner) in a wurtzite-AlN crystal as viewed along the  $[11\bar{2}0]$  direction. The red spots indicate positions of the Al atoms. Evidence of the nitrogen atoms is evident in dimmer shapes, as indicated in the cartoon in the upper left corner.*



In addition to directly imaging the dopant atoms in this crystal, the electron microscope has also been used to locally excite diffusion of Ce dopants that are large compared to Al and N, and Mn dopants that are smaller than both Al and N. Through identifying different diffusion mechanisms, it was found that there is a higher frequency of dopant jumps for the larger and heavier Ce atoms than the smaller Mn atoms. These observations directly confirm density-functional-theory-based predictions of a decrease in diffusion barrier for large substitutional atoms.

R. Ishikawa, R. Mishra, A. R. Lupini, S. D. Findlay, T. Taniguchi, S. T. Pantelides, S. J. Pennycook, "Direct Observation of Dopant Atom Diffusion in a Bulk Semiconductor Crystal Enhanced by a Large Size Mismatch," *Phys. Rev. Lett.*, **113**, 155501 (2014).

## Revealing Buried Quantum Dot Structures in 3-D: Electron Tomography in the Scanning Transmission Electron Microscope

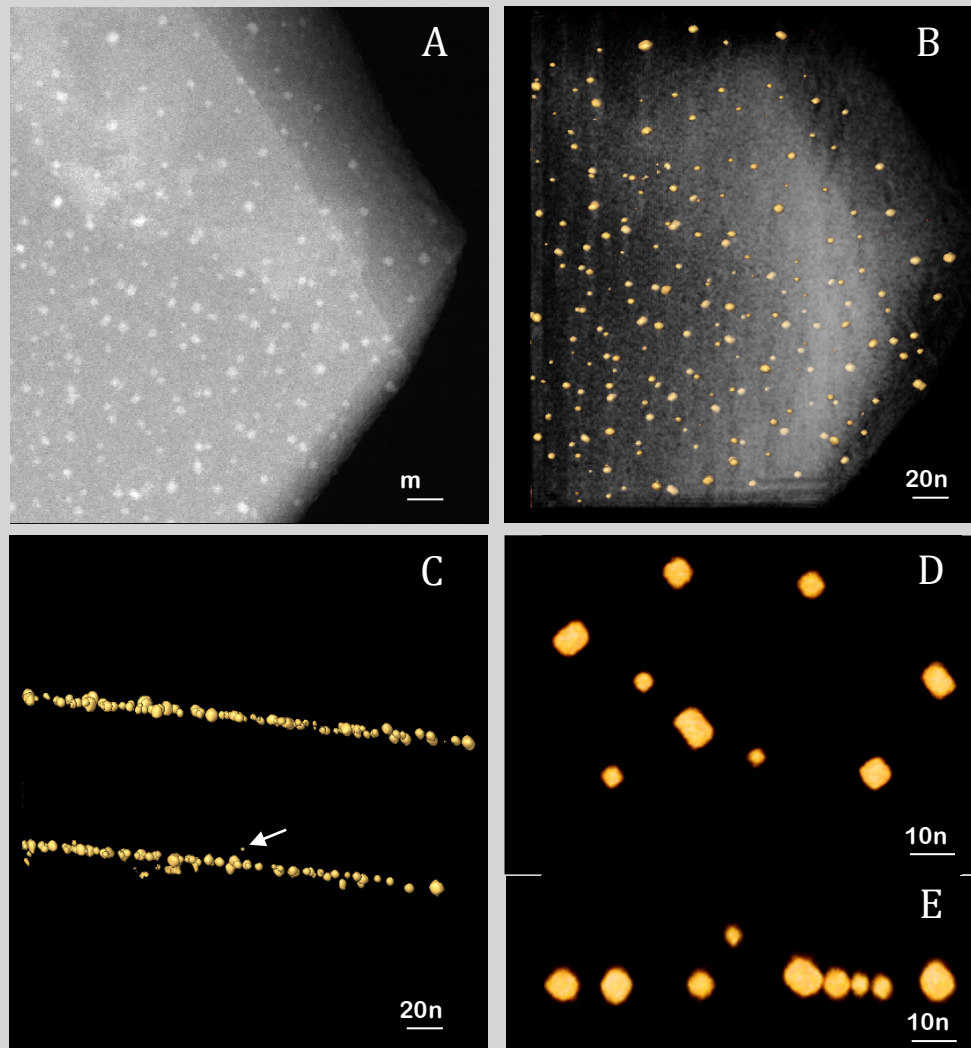
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Electron tomography in the scanning transmission electron microscope (STEM) is one method that is capable of providing the three-dimensional (3-D) structure of nanomaterials. Electron tomography entails taking a series of 2-D images (70-140 images) at many different angles in the electron microscope of the same object, and using various algorithms to reconstruct a 3-D tomogram of the nanomaterial. Over the last 10 years, TEM holder technologies and geometries have improved to provide higher tilt ranges allowing for higher 3-D resolution, while automated acquisition software enables reduced electron beam exposure; and most recently, novel reconstruction algorithms that can drastically reduce the number of images necessary for a high-resolution reconstruction with resolution down to the atomic scale. These advancements in STEM tomography make this technique ripe for the understanding of complex and sensitive qubit structures in 3-D.

One example of a high-resolution reconstruction is a sample grown by molecular beam epitaxy (MBE) that contains a series of Sn quantum dots spaced by layers of Si. The reconstruction can be seen to reproduce the Sn quantum dots' locations and sizes. Note that the specimen was prepared in plan view geometry so that information on the quantum dots in the perpendicular direction cannot be obtained by simply tilting the sample in the

electron microscope. However, after imaging this sample 149 times at different tilt angles, the 3-D reconstruction allows the sample to be viewed in the perpendicular direction. With the exception of a very few quantum dots, the Sn quantum dots are all positioned in two distinct layers. These few dots that are formed outside of the layers are of most interest, as they occur in positions where no Sn was deposited. To determine how the quantum dots form outside of the layers, a reconstruction is performed on the specific volume surrounding the dots of interest. The results indicate that the dots form through Stranski-Krastanov growth from diffusion of Sn in the layers to voids in the Si outside of the layers.

With the synthesis of complex nanomaterials and design of nanostructure devices comes the need to directly visualize the systems in order to ascertain the structure-property relationships that would lead to the next generation of devices. This comparison between quantum dots formed inside and outside the layer of deposition shows that STEM tomography is a valuable technique capable of providing information on the 3-D formation mechanism of nanoscale objects which may be of value verifying emerging fabrication techniques and methodologies used to make next-generation qubit devices.



(A) One of the 149 Z-contrast images acquired at a  $0^\circ$  tilt. (B) 3-D reconstruction of the whole volume showing the quantum dots in yellow and the Si matrix in white from a similar perspective. (C) Cross section of the reconstructed figure with the Si contribution hidden, showing that the Sn quantum dots are mostly contained in two layers, and that there is one small dot in between the layers, as indicated by the arrow. (D) Higher resolution reconstruction of the volume around the out-of-layer dot in  $\langle 100 \rangle$  and (E)  $\langle 110 \rangle$  orientations. Clear faceting reveals the size, shape, and distribution of these quantum dots.

I. Arslan, T. J. V. Yates, N. D. Browning, P. A. Midgley, "Embedded nanostructures revealed in three dimensions," *Science* **309**, 2195-2198 (2005).



## Connecting Atomic Fundamentals to Qubits Through Mesoscale Modeling

Mesoscale approaches allow device-scale models and measurement analytics to leverage first principles atomic-scale simulations. Refined multiscale techniques that preserve quantum characteristics across time, length, and energy scales are essential to predicting the local qubit environment and pathways of information flow and loss.

### **Opportunity**

Complex materials systems, such as those found in quantum computing devices, often exhibit mesoscale phenomena that are not well understood. The mesoscale is defined as the regime where functionality that is critical to macroscopic behavior begins to manifest itself. This is not necessarily at the atomic or nanoscale, but at the scale where defects, interfaces, and non-equilibrium structures are the norm.<sup>21</sup>

Such systems are subject to a variety of time-dependent perturbations and structural inhomogeneities that prevent their treatment as continuum materials. For example, the presence of interfaces and defects—whether designed or unintentional—can significantly change the properties of the material. Poorly understood changes in functionality can, in turn, alter quantum computational device performance in unpredictable ways.

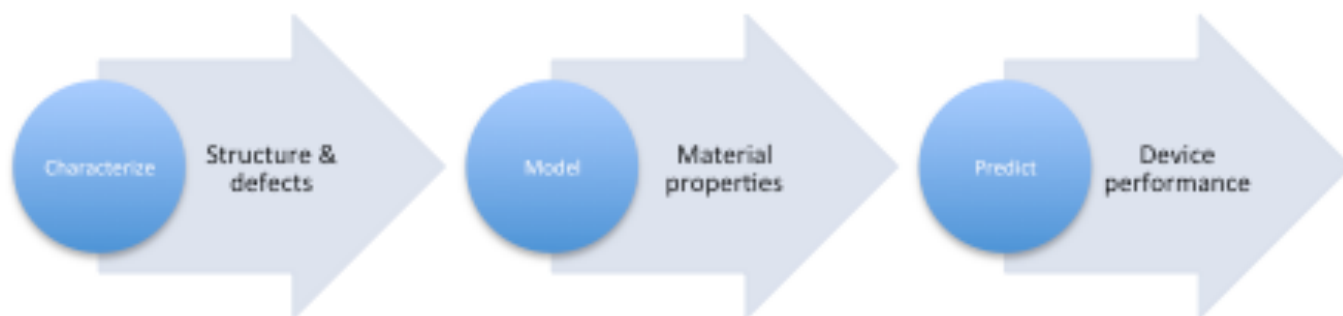
Quantum computation requires precise control of material structure and composition to achieve the fidelity necessary for robust computing. The level of control for quantum computing platforms is significantly more demanding than for current CMOS platforms. For example, some estimates indicate that a 1000-fold increase in control of the electronic

structure may be required (meV to  $\mu$ eV). Such control will necessitate much greater spatial and temporal resolution in materials fabrication, composition, and characterization.

The influence of disorder and noise such as electronic and structural defects, environmental field noise, and time-dependent fluctuations must be understood in order to design materials that can meet the tight performance requirements for quantum computing. However, the current state of the art lacks the theoretical, mathematical, and computational capabilities to directly connect materials properties resulting from material synthesis and characterization to predictions about quantum device performance. There is a critical gap in mesoscale modeling in the make-measure-model design cycle for quantum computing materials.

### **Timeliness**

The foundation for bridging this capability gap exists and has been highlighted by the DOE. With knowledge of the atomic and nanoscale behavior of materials, the study of more complex systems is possible. Advances in computational capabilities provide strategic positioning that can be used to unravel and control the complexity that determines functionality at the mesoscale<sup>21</sup>. These rules, and their application to quantum materials, are being explored at multiple DOE labs and in academic research groups. While investments in studying mesoscale materials are providing an essential foundation for quantum computing materials research, they are not specifically focused on quantum computing and additional advances are needed to make these approaches readily applicable to quantum computing materials.



*Illustration of connections between physical structure of a material and device performance made possible through mesoscale computational techniques (Baker 2014).*

### **Approach**

Mesoscale modeling approaches can bridge scales from electronic and atomistic to devices. The concept of implementing a computational/experimental cycle into the discovery and development effort is well known in mesoscale systems for energy applications. For quantum computing technologies, computational tools that propagate atomistic and electronic information across multiple length scales into a device-scale description of qubits, while accounting for and/or providing estimates of the errors, are critically needed to enable the design and optimization of operational qubit devices. Current modeling tools can typically best address either short- or long-range interactions but are not able to accurately integrate across these scales. The self-consistent coupling between short- and long-range interactions is critical to accurately simulate how a qubit interacts with its environment and how materials that constitute that environment can be better tailored to enable longer coherence times and enhanced ability to probe qubits. These interactions are anisotropic and involve active interfaces and surfaces, and thus are fundamentally important to controlling the function of an operating device. In order to model these interactions accurately, we need new multiscale algorithms and solvers that can account for local interactions at the

material interfaces, while including the mean-field effects of long-range interactions with the bulk<sup>22,23</sup>. In most qubit systems, the algorithms need to incorporate time dependence, temperature dependence, and externally driven sources through a time-dependent term in the Hamiltonian of the system. This requires building the underlying algorithms that couple ab initio molecular dynamics and mean-field (macroscopic) governing equations to actively explore important science questions such as qubit fidelity, how to control stray electric fields in the materials, and how to maintain qubit entanglement in an open environment.

Coarse-graining approaches connect atomic models to continuum scales by systematically reducing the degrees of freedom. Understanding which atomic-scale materials properties are central to qubit system performance (and which are not) is a key challenge. Spatial coarse-graining can significantly improve simulation speeds and enable the connection of length scales to address various qubit-qubit interactions; however, care must be taken to also accurately capture the dynamic behavior of the coarse-grained system and the propagation and compounding effects of errors and uncertainty. The simple Langevin or Brownian stochastic methods used with many coarse-grained models are often unable to reproduce important system properties

such as memory effects or hysteresis. Instead, more sophisticated dynamics methods such as generalized Langevin or Mori-Zwanzig approaches must be used in conjunction with spatial coarse-graining to provide faithful representations of system dynamics. Here, care must be taken during coarse-graining so that the parameterization of noise and materials models based on atomistic-scale results are in a form that can be explicitly or implicitly fed into a device model.

Finally, concurrent coupling approaches can incorporate detailed quantum mechanical descriptions of site defects and inclusions into coarse-grained descriptions of the larger-scale material environment. Concurrent coupling methods provide a framework that allows multiscale interaction between models. This interaction goes beyond conventional bottom-up coarse-graining by allowing large-scale macroscopic behavior in the system to influence the fine-scale atomic simulations. Such approaches have previously been successfully applied to integrate atomic-scale information into continuum models of materials mechanics. The integration of quantum mechanical models into a concurrent coupling framework will provide new mathematical challenges, but it is an important component

for more realistic modeling of quantum computing materials and the coupling of their associated electron and spin degrees of freedom with external noise sources.

Many of these mesoscale methods will be computationally expensive and therefore must directly leverage DOE investments in scalable solvers and leadership-class computing platforms. The coupling from nanoscale through to device-scale will also require experiments designed to specifically address issues underpinning the coupling between different models that are appropriate to different scales.

### ***Potential Impact***

Significant modeling advances are required to better understand the atomic origins of quantum computing materials properties. However, such advances offer the promise of tremendous impact on quantum computing by enabling the make-measure-model cycle that is essential to rational materials design. A specific near-term impact will be an increased ability to understand and harness disorder and noise for improved robustness in quantum devices and functional materials more broadly.

## Mesoscale Modeling for Charge Noise in Silicon Qubit Devices

Many important properties of the local environment that impact qubit performance are fundamentally mesoscale in nature. Mesoscale modeling involves a range of mathematical and computational challenges that promise direct improvements in the characterization and design of quantum-grade materials. For example, charge disorder arises in highly localized regions of materials but can have a significant impact on the macroscopic current-voltage behavior in qubit systems. Detailed modeling of material structure and the influence of defects and disorder on larger-scale behavior has several potential benefits, including the ability to more accurately describe the perturbation of system energy valleys due to their presence. Charge disorder is a relatively common type of defect that can disrupt the device barriers needed to localize charge and create roughness in the local potential that can interfere with charge localization.

As illustrated in Figure 1, computational and mathematical foundations currently exist for investigating phenomena at the atomic and continuum scales but additional research is needed to connect these scales. For the moment, consider how mesoscale modeling can positively impact silicon-based quantum computing.

**At the atomic scale,** quantum mechanical density functional theory (DFT) calculations have been used to explore paramagnetic noise in aluminum oxide quantum computing materials as well as charge carrier interaction with defects in aluminum antimonide semiconductors. In amorphous silicon oxide, the primary type of defect is an E' center, where an O<sup>-</sup> anion is removed from the system, leaving a Si<sup>+</sup> site and electron in the system. Although these types of charge defects are

fairly dilute, their impact on system behavior can be significant and is poorly understood. Development of quantum Monte Carlo methods for defects may provide an excellent path toward deriving and systematically improving appropriate density functionals for input to the DFT. DFT methods themselves provide an excellent framework for exploring the fine-scale influence of such defects; but need to be connected with continuum models of device behavior in order to realize their predictive capabilities for quantum computing. Quantum transport from the qubit to the continuum measurement world will also require implementation methods that are specific to the exact material realization of the qubits.

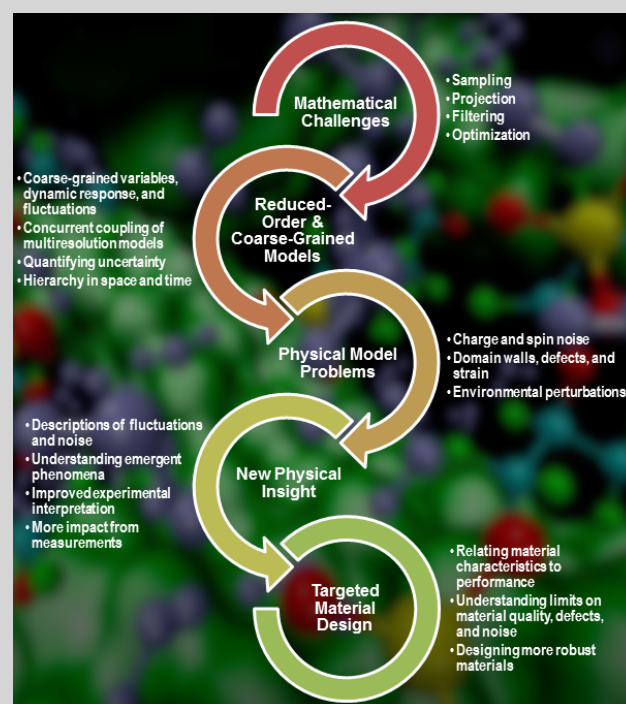
**At the continuum scale,** semi-classical Schrodinger-Poisson finite element solvers have been used for device modeling and offer broad optimization capabilities and the ability to perform *some* design iterations of quantum computing devices, such as investigating S-dot-D tunnel barriers. QCAD is an open-source tool developed to use these methods for the design and analysis of silicon-based quantum computing devices. However, current methods for modeling charge disorder and charge noise via QCAD use over-simplified models of charge density spread uniformly throughout the oxide layer of the device. More sophisticated DFT descriptions are needed to provide more accurate models of charge defects and provide predictive power for understanding device characteristics. However, the current continuum framework used by QCAD is unable to incorporate such information at length- and time-scales relevant to device behavior.

**Mesoscale methods** can be used to relate the results of atomic-scale methods such as DFT to macroscopic quantities of interest in the

context of QCAD. For example, insight from quantum mechanical models into the nature and distribution of E'-center defects can be incorporated into continuum models to understand the influence of these defects on device properties such as current-voltage relationships.

Uncertainty quantification (UQ) approaches provide a straightforward path for incorporating more detailed charge noise distributions *and* examining the influence of these distributions on device property quantities of interest. Non-intrusive “black box” UQ methods, such as stochastic collocation, offer a straightforward method for incorporating small numbers of point-like charge defects into the modeling software, such as QCAD, with no modification of the code. However, more efficient and complete characterization of defects on device performance requires the application of more sophisticated UQ methods such as generalized polynomial chaos and stochastic Galerkin methods. While harder to implement, such methods allow more general charge disorder distributions and provide more efficient analyses of the effects of charge noise on quantities of interest.

Ultimately, the highest levels of accuracy and prediction will arise from direct coupling of atomic-scale DFT models of defects with QCAD-like device models. Such coupling is important as it not only allows detailed integration of defect charge distributions into device-level models but it also allows macroscale device behavior to perturb the electronic behavior at the defect site. Concurrent coupling methods provide a framework that allows such multiscale interaction between models. Such approaches have previously been successfully applied to integrate atomic-scale information into continuum models of materials mechanics. While the integration of quantum mechanical models will provide new mathematical challenges, this previous work lays a foundation for true mesoscale modeling of materials for quantum computing.



**Figure 1.** An overview of mesoscale modeling challenges in quantum materials characterization and design.

## Revealing Qubit Dynamics and Decoherence Mechanisms

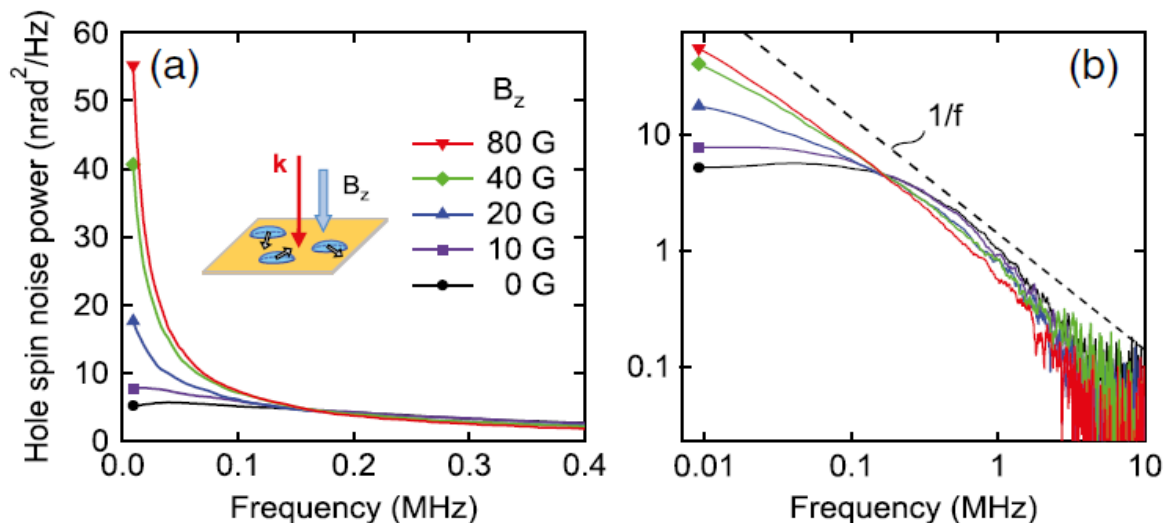
The interrogation of ground state and near-ground state qubit interactions with the local charge and spin environment is a critical evaluation accomplished for qubit devices. Both passive and perturbative techniques that assess these characteristics on multiple length scales and qubit-relevant timescales without fabricating qubit devices offer clear pathways to accelerating control of qubit materials.

### Opportunity

Interrogation of atomic, charge, and spin systems is vital to the advancement of quantum computing materials. The techniques used can be either passive or perturbative, employing photons with energies that are resonant with the energies in the operating regimes of the qubit, resonant with the fundamental atomic structure of the quantum device, or non-resonant. All energy and time scales of photon interactions with quantum computing

devices offer potential metrological insight that may provide new opportunities to learn about the local quantum environment experienced by the qubit.

An important measure of the performance of any qubit is the coherence time that limits the number of operations performed without introducing significant error to information flow. This parameter is ultimately limited by qubit materials properties, which are further degraded by subsequent imperfect synthesis and fabrication processes. Understanding the mechanisms that are responsible for qubit loss and decoherence is an essential and challenging task since material functionality emerges from strong interactions among spin, lattice, charge, and orbital degrees of freedom that are hard to separate with conventional experimental approaches. Another important aspect of qubit material physics is the dynamic behavior, including the identification of fundamental timescales of the qubit interactions with the local electronic, structural, and magnetic transformations in the material.



*Spin noise of hole spin qubits in InGaAs quantum dots, on linear and log-log scales (left/right panels). The noise evolves from Lorentzian to power law scaling, indicating a change of the underlying decoherence dynamics. (Zapasskii 2013)*



Passive or “weak measurements” allow one to probe the quantum state of a system and gain insight into its Hamiltonian or the respective density matrix, while leaving the quantum system nearly unperturbed. Perturbative or non-equilibrium techniques allow characterization and manipulation of materials properties at the fundamental time and space scales of atomic and electronic motion. Femtosecond temporal resolution and broadband spectral selectivity enable studies of charge, spin, and lattice dynamics, and more importantly, provide a means for temporal discrimination of coupling between these excitations. Recent developments in nonlinear optical techniques have extended ultrafast measurements to both soft x-ray and terahertz frequency regimes. Analogous measurements, albeit on longer timescales, in the microwave regime are commonly used for qubit manipulation and readout.

### ***Timeliness***

The dynamical properties of electron and hole spins in bulk and quantum confined nanostructures are actively being investigated for potential applications in quantum information processing. Optical pump-probe studies based on ultrafast techniques have proven essential in this regard, directly revealing the g-factors and coherence decay processes of spins in these materials. Recently, these important properties have been shown to be accessible via alternative measurement approaches based on spin noise spectroscopy (SNS), in which the intrinsic fluctuation spectra of the spins also reveal this dynamical information.

Such a noise-based approach to spin resonance is in marked contrast to conventional nuclear- or electron-spin resonance techniques, which necessarily perturb the spin system. Because a specific external stimulus is not required to produce a signal, SNS is well suited to the detection of

previously unanticipated noise sources and unknown mechanisms. The fluctuation-dissipation theorem underpins this noise-based approach, and proof-of-principle measurements already have been demonstrated. Importantly, noise-based measurements become increasingly attractive alternatives to conventional dissipative methods as the few-particle limit is approached, since noise signals fall only as the square root of the number of noise sources rather than scaling linearly as in other regimes. It is worth noting that the power and utility of noise-based techniques are gaining significant appreciation in many related disciplines and may find direct application in quantum computing devices.

Currently, the well-established methods of x-ray imaging and spectroscopy are moving to the attosecond domain and enable direct insight through time-resolved “movies” into the nanoscale dynamics using different contrast modalities sensitive to chemical, strain, magnetic, and electronic features. Similarly, intense ultrashort mid-infrared and terahertz (THz) pulses can now be used to directly photoexcite and probe specific low energy modes, such as soft mode phonons and magnons, improving upon previous all-optical techniques that only indirectly couple to these excitations. This offers an exciting opportunity for coherent manipulation of any imaginable excitations relevant to qubit operation at timescales inaccessible by current electronic approaches. Novel large-scale and tabletop coherent sources of intense sub-picosecond THz and x-ray pulses along with novel spectroscopic and imaging techniques are beginning to allow direct probing and manipulation of spin, lattice, and charge dynamics, along with in situ imaging of dynamic processes occurring at the nanoscale.

## **Approach**

Recently developed methods for optical SNS may provide a viable method for noninvasively probing spin systems by using detuned probe photons to couple to a qubit's spin degrees of freedom. These studies are directly relevant to the burgeoning field of quantum measurement science.

Not all noise in experimental measurements is unwelcome. Certain fundamental noise sources contain extremely valuable information about the system itself. A classic example is the inherent voltage fluctuations that exist across any resistor (Johnson noise) from which temperature can be determined. In spin systems, fundamental noise exists in the form of small, random spin fluctuations. For example, statistical fluctuations of paramagnetic spins should generate spin noise even in zero magnetic field, thus perhaps enabling a new method to evaluate material defects before fabricating qubit devices. Crucially, the frequency spectra of measureable fluctuations reveal characteristic dynamic properties of the spins, such as spin precession frequencies, decoherence times, and inter-particle couplings, that may enable categorizing noise sources without ever having to perturb the spin ensemble away from thermal equilibrium.

Application of established techniques and the development of new techniques in optimization and machine learning will help advance the interpretation of these SNS measurements to systems with smaller spin populations. More advanced methods often couple techniques arising from time series analysis and reinforcement learning to gain insight into the structure of unknown functions. Entropy-based methods provide a natural language to pursue optimization in the context of SNS. Not only are physical systems naturally governed by entropy, but

entropy methods employ mathematical formulations which have been applied in signal processing to enhance and extract features.

Perturbative techniques such as ultrafast optical spectroscopy (UOS) complement passive methods. UOS takes advantage of the fact that characteristic timescales of dynamics of various degrees of freedom (DOF) differ by several orders of magnitude from a few to tens of femtoseconds for electrons to one to a few picoseconds (ps) for phonons to tens to hundreds of ps for spins. This difference allows their discrimination in the time domain. Typical UOS measurements consist of sending an intense pump pulse to create an initially non-thermal electron distribution that alters the response measured by a time-delayed probe pulse. During the first  $\sim 100$  femtoseconds (fs), the non-thermal distribution generally relaxes by electron-electron scattering. Subsequently, the hot Fermi-Dirac distribution thermalizes through coupling to other DOF such as through phonon or magnon emission or polaron redressing. The sub-10 fs temporal resolution and spectral selectivity available with UOS enable reliable separation of these dynamics and can thus reveal the DOF and interactions involved in material functionality as the system returns to equilibrium. In addition, recent UOS advances now allow complete control over the phase, amplitude, and polarization of laser pulses with durations well below a multitude of dephasing times ( $\sim 100$  fs). This provides means to guide a system along particular quantum pathways and study the effects of various materials and environmental parameters on qubit coherence. In particular, polarization-controlled pulses can be used to coherently manipulate spin alignment on femtosecond timescales.<sup>14</sup>

All optical UOS methods rely on high-energy (1-4 eV) photons that are perturbative and

often probe materials properties very far from the ground state. Alternatively, terahertz spectroscopy uses low-energy (1 THz  $\sim$  4 meV) photons to probe charge dynamics near the Fermi energy. In addition, the fields of single-cycle THz pulses transmitted through the sample can be detected coherently, thus allowing the complex conductivity and permittivity to be extracted without the complex analysis required for visible photons. Arguably, the most promising application of THz pulses is to coherently drive low-energy excitations (magnons, phonons, etc.) and study their effect on the functionality of qubit materials. These pulses can be used to coherently manipulate spin, strain, or other relevant qubit states and probe decoherence pathways and timescales with femtosecond resolution.

The next generation of qubits will have buried interfaces that will require probes with deep penetrating capability and simultaneous nanometer and femtosecond resolution. The recent extension of the well-established methods of x-ray absorption/photoemission spectroscopy and coherent diffractive imaging to the femtosecond domain has enabled researchers to directly probe lattice, spin, and charge dynamics with high elemental specificity and sensitivity to buried interfaces. In particular, time-resolved x-ray magnetic circular and linear absorption dichroism (tr-XMCD/XMLD) spectroscopies provide a means to measure the degree of ferromagnetic and anti-ferromagnetic spin alignment in the materials by probing the polarization dependence of the x-ray absorption at element specific resonances. This can now be accomplished with tabletop tr-XMCD/XMLD spectroscopic capabilities that provide direct access to photo-induced spin dynamics with sub-50 fs temporal resolution. A similar x-ray source can also be used to produce photons for time-resolved

angle resolved photoemission spectroscopy (tr-ARPES), which provides a unique ability to measure the excited state evolution in energy and k-space near the surface. Both techniques enable the investigation of spin, lattice and charge dynamics that underpin decoherence and energy transfer pathways in prospective qubit materials.

### **Potential Impact**

Spin noise spectroscopy enables the measurement of the dynamic properties of spins relevant for the development and optimization of materials for quantum information applications, including spin precession frequencies, decoherence times, and inter-particle couplings. Crucially, these noise-based measurements are largely passive and non-perturbing, and in many cases can be considered true “weak measurements.” When combined with powerful new methods of computational analysis and optimization, this technique will enable an understanding of the noise processes underlying decoherence in a quantum system but will also provide understanding of the underpinning for the development of devices for spintronics.

Despite rapid progress in ultrafast photon source development and their application in understanding materials functionality, novel UOS techniques have not yet been used to study or control qubit dynamics. UOS enables dynamics investigations covering a broad spectral range, spanning x-rays to millimeter waves, and therefore has the potential to enable significant advances in not only our understanding of material and qubit behaviors, but also our ability to control its state and coupling to various environments. Therefore, the impact of applying and advancing these techniques will reach far beyond quantum computing, as the experimental and theoretical knowledge base developed during qubit/material exploration



*Advanced Photon Source at Argonne National Laboratory provides a promising avenue of investigating materials for quantum computing research.*  
 ([http://chemgroups.northwestern.edu/chen\\_group/facilities.htm](http://chemgroups.northwestern.edu/chen_group/facilities.htm))

at extreme time and spatial scales could be used to push the advancement of the material science in a broad context.

### **Cultivating the Precise Fabrication of Ultrapure Qubit Devices**

Quantum computing technologies have requirements for device features and purity that are more technologically demanding than for established industries. Turning quantum-grade materials into reliable qubit devices requires advancements in existing fabrication tools and techniques as well as the development of new approaches to enhance our ability to control quantum materials. Correspondingly, meeting the challenges of quantum computing enables a new era of synthetic control in general.

#### **Opportunity**

The quantum nature of the qubit causes sensitivity to the nanoscale and mesoscale

environment that are intimately related to the materials from which the devices are made. Polycrystalline, nanocrystalline, and amorphous materials have several critical roles. For example, consider insulators in semiconductor qubit technologies, which are used for gate oxides to form high resistivity electrical point contacts. These contacts are needed for control of the local potential in silicon germanium quantum dot and donor dot devices. Charge noise resulting from defects in these materials or at the interfaces between the oxide and metal, or oxide and semiconductor limits device performance. Conductors are needed for electrodes that bias devices into operational regimes across all technologies. The purity, stability, and homogeneity are all concerns for these materials as any fluctuation in the local conductivity inherently leads to variation in the local charge environment.

Amorphous  $\text{Al}_2\text{O}_3$  is ubiquitously used in the formation of superconducting Josephson junctions (JJ). Today's highest performing

superconducting quantum computing devices are made from sub-micron area JJs using a trilayer of aluminum, aluminum oxide, and aluminum via shadow evaporation techniques and electron beam lithography. The small areas are an engineering solution that minimizes level splittings observed in the spectroscopic analysis of qubits. These level splittings do not necessarily limit the coherence times of these devices, but they do dramatically limit the reliability of measured devices, which is a challenge for experiments that rely on entangled qubits. The origins of these level crossings are poorly understood, but it is likely that the non-uniformities of the oxide layer from either individual point defects or simple inhomogeneities of the amorphous material combine to limit performance of large junction area devices. This issue offers opportunities for characterization and process improvements that could rapidly impact the reliability of Josephson junction-based quantum computing devices.

Lithographic processes with unprecedented resolution are also required to form the most reliable superconducting Josephson junction circuit elements, define the nanometer scale electrodes required for semiconductor based quantum computing, and position single dopant atoms in precise locations for donor-based silicon quantum computing technologies. The accuracy and precision requirements for superconducting quantum computing is comparable to large-scale CMOS capabilities, while semiconductor quantum computing technologies are beyond the capabilities of modern day CMOS tools.

### ***Timeliness***

Recent laboratory-level demonstrations of near atomic-precision lithographic capabilities provide a potential path towards an idealized bottom-up, atom-by-atom, device fabrication process<sup>24</sup>. Scanning

tunneling microscopes (STMs) and helium ion microscopes (HeIMs) are tools typically used for analyzing surfaces. However, the highly focused nature of their respective probes (tunneling electrons for STM and accelerated helium ions for HeIM) is ideal for manipulating materials at atomic length scales to fabricate quantum computing devices through lithographic processing.

A well-defined process is established for the precise placement of phosphorous donors in silicon to fabricate single qubit devices with STM-based hydrogen lithography. This process was initially used to fabricate a number of “rudimentary” devices and structures such as atomic wires, single atom transistors, charge sensors, and in plane gating, which led to more complex devices for donor spin readout. The current status of the process has many challenges associated with pattern reliability and chemical process control that presently hinder the ability to fabricate devices on a regular basis. However, enough information can be extracted from prior investigations to parse these challenges into manageable research topics upon which to focus future efforts.

The helium ion microscope has grown out of the development of the scanning electron microscope, which is reaching its theoretical limits of performance stemming from the nature of its source. Even though the helium ion microscope is based on more than half a century of work in field ion microscopy and scanning electron microscopy, the first helium ion microscope was only delivered to the National Institute of Standards and Technology in 2007. The current resolution of helium ion microscopes is limited by the ability to produce stable ion beams at the required energies. A potential enhancement of helium ion microscopes for lithographic applications is the replacement of helium with neon as the gas field ion source.

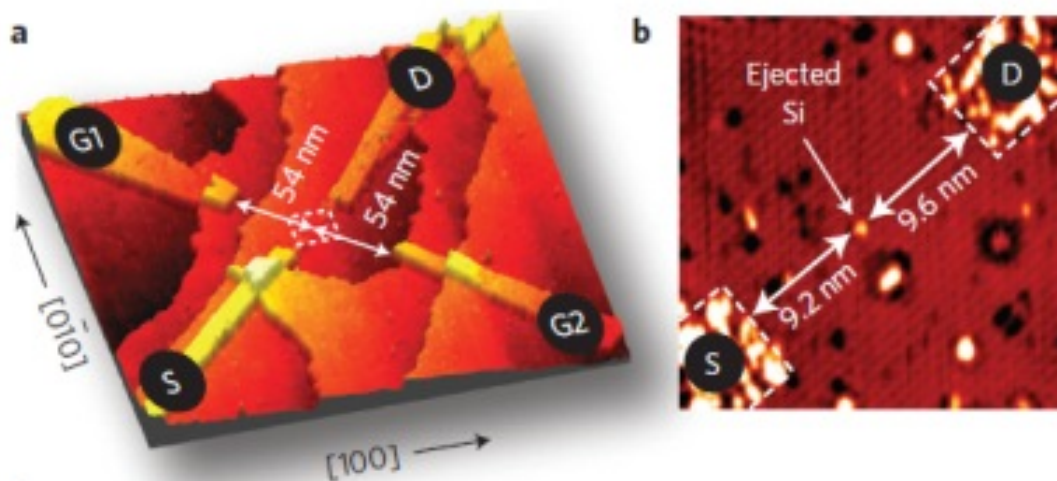


## Approach

Improvements in clean room processing are an area where existing technologies can be leveraged and extended to enable the repeatable fabrication of high-quality materials. Although different processing procedures are often tested and optimized by individual research groups, the direct connections of a procedure to materials properties are not readily available. This currently unexplored area that correlates processing procedure with device performance can be addressed through more complete materials characterization following the use of specific tools and processing steps. For example, enhanced homogeneity for amorphous materials and microstructure engineering for contact lines could improve qubit performance over that resulting from the nanocrystalline metals

as material probes, incorporating extensive materials characterization.

STM is a diverse technique that permits imaging, spectroscopy, and atomic manipulation for a number of applications including quantum computing. The leading approach to fabricate semiconductor donor-based qubits is STM-based hydrogen lithography, in which the scanning tunneling microscope is used to pattern a hydrogen-terminated silicon surface, defining locations for phosphorous dopants with atomic precision. Despite a demonstrated ability for atomic precision device fabrication, many challenges exist to make this a viable technique for fabricating larger, more complex device structures, including improvements in the accuracy and reproducibility of lithographically defined patterns and enhanced control of chemical



*A single electron transistor fabricated from metal contacts and a single phosphorous atom at two scales. The device is fabricated and imaged using scanning tunneling microscopy. (Fuechsle 20012)*

that are current employed. Further, exploring not only optimized structures, but also the correlation between the processing procedure, device performance, and failure analysis will be essential, and thus drives a comprehensive approach that requires qubits

processes associated with dopant placement and incorporation.

The accuracy and reproducibility of lithographically defined patterns is highly dependent upon the reliability of the STM probe tip, which is typically derived from its



sharpness and robustness towards the extremely high electric fields (on the order of  $10^7$  V/cm) that it experiences. A vast amount of literature exists on the fabrication of sharp probes for STM, but it is still very much an art form as opposed to a science. Better characterization of the various methods is necessary to properly quantify the resultant tip shape as well as the choice of tip material. Further challenges to accuracy and reproducibility are attributed to the performance of the STM piezoelectric scan tube that underlies the motion of the STM probe tip.

To improve devices currently made at the resolution of electron beam lithographic techniques, the helium ion microscope may be a critical technology. In conventional scanning electron microscopes, the diameter of the electron beam limits performance. The intrinsically shorter wavelength of the incident beam in helium ion microscopes should permit subatomic characterization and nanofabrication. While helium ion beams have induced damage, creating lattice vacancies and interstitial atoms, their increased resolution may prove to be useful. Finally, other emerging techniques such as dip pen nanolithography and field enhanced oxidation, which locally add material have also demonstrated patterning capabilities at length scales below 10 nm, and may be effectively applied to fabricated nanometer scale quantum computing devices. The optimization and application of these techniques to the fabrication of qubits are still in their infancy and require further investigation.

All methods of atomic-scale lithography require enhanced understanding of the surface chemical processes associated with placement and incorporation of the deposited atoms in order to improve control of the overall device fabrication process and maximize yield. Current procedures for

fabricating semiconductor donor dot devices through placement and incorporation of single dopant atoms are complex, involving the adsorption and decomposition of a dopant precursor molecule into various reaction products within a lithographically defined area. Moreover, the local environment surrounding the patterned areas has the potential to alter the incorporation pathway. Investigations of the reaction pathways within idealized lithographically defined patterns as well as the impact of nearby defects and non-idealized lithographically defined patterns will be required to optimize dopant incorporation yield.

To date, investigations into single dopant incorporation have focused exclusively on phosphorous donors, but there is a desire to extend the approach to acceptor dopants. Any extension of the process will require the identification of an appropriate combination of dopant precursor molecule and resist material, as well as establishing well-defined reaction pathways ultimately leading towards single dopant incorporation.

Device structures from all quantum computing devices may benefit from future advancements in nanolithographic processing. Design improvements of the device fabrication processes and process flow may offer to mitigate the technological limitations imposed by the current designs or allow materials to be processed in a manner that improves their realized performance. The fabrication of JJ devices that do not require large resist undercuts, thus removing the necessity of resist bridges, may improve the final material quality of the completed junction. Enhanced lithographic resolution may also impact semiconductor quantum dot devices by improving the dielectric quality and design of contacts that may help reduce cross talk and gate leakage.

## Potential Impact

Quantum computing technologies require device features that are created on the atomic scale and fabricated with exacting purity. Emerging lithographic techniques for qubit fabrication with atomic-scale resolution such as scanning probe microscopy and helium ion

microscopy will also continue to advance imaging, nanofabrication, and analyses that may be leveraged across the physical and biological sciences.

## High-resolution On-demand Patterning

The atomic resolution provided by scanning probes has generated a number of approaches toward building nanoscale materials by design. Better known methods include dip pen nanolithography and field enhanced oxidation, which locally add material, and mechanical indentation or electrochemistry, which can remove material. Each of these has been demonstrated at length scales below 10 nm.

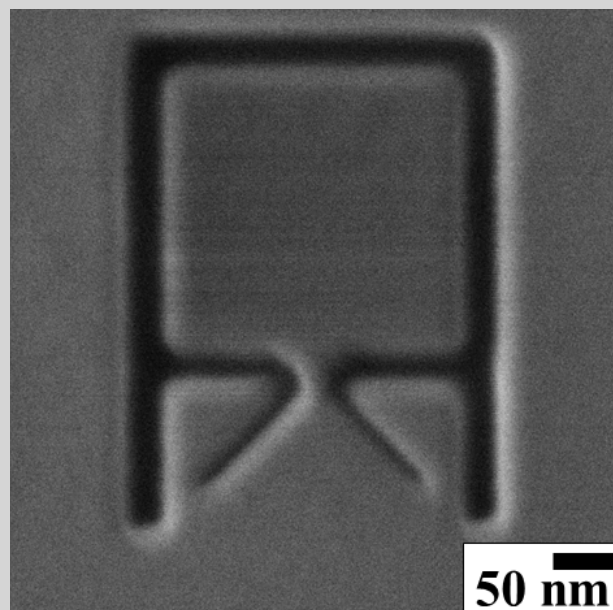
The leading approach of semiconductor donor qubits is scanning tunneling lithographic microscopy where the scanning tunneling microscope is used to pattern a hydrogen terminated silicon surface defining locations for phosphorous doping, with atomic precision, that is then overgrown to create a three-dimensional structure. From an abstract level, this point-by-point determination of phosphorous atoms in a three-dimensional structure is similar to additive material fabrication where control of donor atom placement could be completed from a series of CAD drawings describing each atomic layer.

The helium ion microscope (HeIM) enables lithographic strategies that can help realize advanced qubit architectures without the limitations of traditional optical or electron beam techniques. (Reference: Joy, D. C. (2013). *Helium Ion Microscopy Principles and*

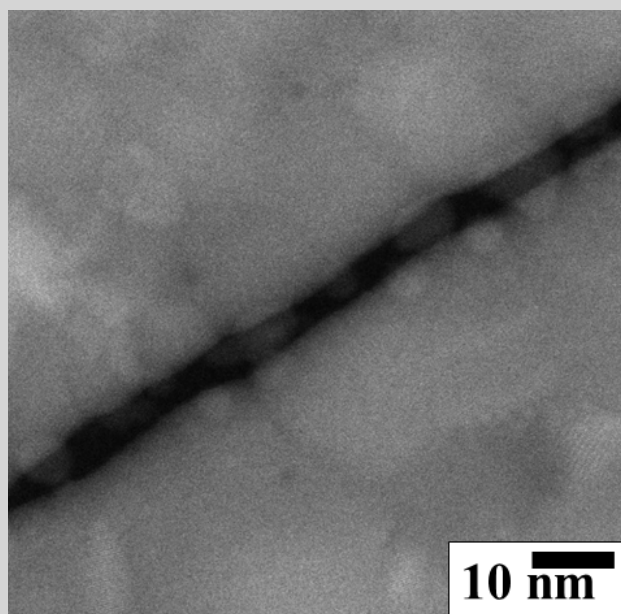
*Applications*. New York. Springer.) The HeIM's 0.5-nm probe and pattern generator enable the ion beam to expose or mill complex patterns in a variety of materials, with or without lithographic resists, achieving better resolution than electron systems due to a lack of proximity effects and a comparatively small interaction volume. Feature sizes approaching 10 nm as illustrated in Figure 1 of a diode test structure patterned resist-free from single-layer graphene on an insulator have been achieved. Also HeIM direct write has achieved a 1-nm line width as confirmed in the transmission electron micrograph (TEM) in Figure 2 illustrated by a HeIM-patterned line on a silicon nitride TEM grid.

Further, equipping the HeIM with a gas injection system enables nanoscale additive techniques: material can be deposited with nanometer precision, again without the need for masks, by a controlled decomposition of gaseous organometallic precursors by the ion beam. A variety of materials can be direct-deposited by the HeIM including gold, platinum, tungsten, cobalt, and silicon oxide. A combination of HeIM lithographic techniques allows for the construction of complicated circuits and structures without ever exposing the sample to resists, solvents, or etchants.

The HeIM may also be applicable to microanalytical characterization of qubits, which is necessary due to the quantum-grade purity requirements. Further advances in HeIM include proposed methods for microanalysis on the nanometer and possibly subnanometer scale. Microanalytical information will be derived from energy spectra collected by using Rutherford backscatter ions or secondary ion mass spectrometry. The data generated would identify not only elements but compounds as well.



**Figure 1:** Helium ion image of a HeIM-fabricated, resist-free, direct write lithography of electrical pathways in a diode test structure from single layer graphene on silicon insulator.



**Figure 2:** Transmission electron micrograph (TEM) of HeIM direct write on a silicon nitride TEM grid that has been gold-coated to facilitate imaging to confirm 1-nm line width.

## Authenticating Qubit Devices Using Integrated Models

Dramatic improvements in fabricating high-quality qubit devices and interpretation of actual qubit behavior rely on creating a design toolset for quantum circuits. Efforts to improve repeatability and reliability can move beyond correlational approaches to causal design and fabrication workflows by using integrated predictive modeling to connect fabrication with materials, devices, and system performance.

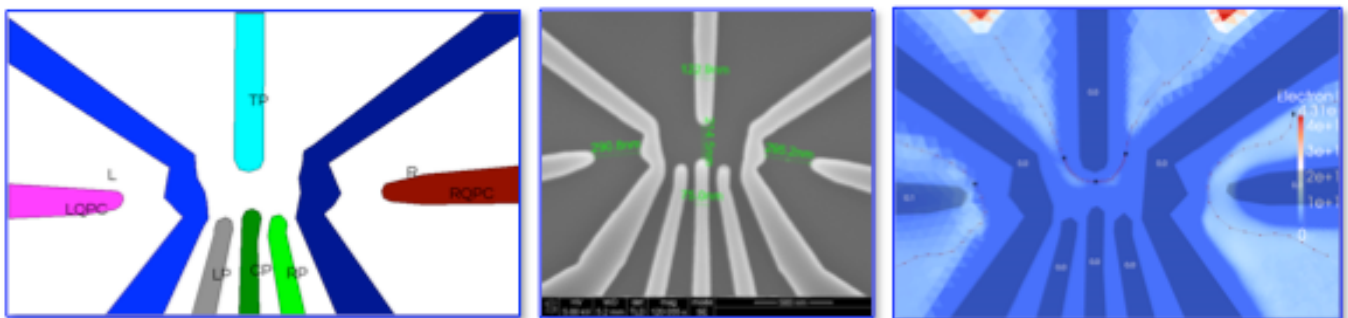
### **Opportunity**

The same physics that allows fundamentally greater computational power to result from a few atoms in quantum devices makes these devices sensitive to defects, disorder, and atomic-level noise in ways that traditional computing devices are not. In particular, the small scale of quantum devices makes them sensitive to disorder on the same length scale. Moreover, the necessity for quantum devices to establish and maintain entanglement renders them sensitive to environmental fluctuators that can become entangled with the desired quantum states. These challenges have slowed the rate of progress in designing high-fidelity quantum devices, and necessitate the development of a complete quantum device modeling capability tightly coupled to experimental characterization and device fabrication, tying

process parameters to the resulting quantum device characteristics so as to create quantum devices robust to defects, disorder, and noise. Finite-element device-level simulations for semiconductor, superconducting, and trapped-ion qubits can predict the response of these devices to a variety of inputs, intended and unintended, and thus present the opportunity to assess process and design parameters in advance of fabrication to understand their impact on qubit performance. Such a capability promises to accelerate the development of multiple qubit technologies as more exquisite control is imparted to fabrication processes to achieve devices with the desired and designed specifications.

### **Timeliness**

Most qubit technologies have reached a critical stage in their development, in that fundamental explorations have been performed, and promising approaches have been identified and are now the target of more focused optimization. Mastery of noise and disorder in these systems through advanced materials modeling and characterization techniques will be an important accelerator of progress to realize the potential of qubits. Furthermore, such mastery will be critical as the field transitions from creating isolated demonstration devices to more reproducible, scalable, and manufacturable systems.



*Three perspectives of a silicon quantum dot device. The mask layout (left), electron image of the fabricated device (center) and simulated potential landscape (right). (Muller 2014)*

## Approach

The approach that the semiconductor industry currently uses to map processing parameters and the results of extensive characterization to device characteristics is a paradigm that needs to be adopted for the optimization of quantum devices. However, quantum devices present additional challenges such as the fact that candidate devices involve single (quasi) particle structures, which means that isolated defects can dramatically affect performance; and the reliance of quantum algorithms on entanglement exposes them to environmental defects that can become entangled with the desired quantum state, creating further decoherence channels. Progress in the development of quantum devices requires advancement of the classical microelectronics process models into the quantum regimes.

The semiconductor industry relies on a variety of fabrication process and device modeling tools that fall under the heading of technology computer-aided design (TCAD). The device modeling tools typically use finite-element modeling (FEM) methods to discretize the Poisson equation describing the electrostatics of semiconductor devices composed of a variety of different materials, with different doping values, and with different boundary conditions arising from voltages applied to gates, ground planes, and the like. Several modifications are required for TCAD tools to describe quantum systems. The electrons in low-temperature quantum devices typically obey Fermi-Dirac statistics rather than Boltzmann statistics, requiring the use of at least a Thomas-Fermi nonlinear Poisson equation to be solved for the electrostatics, or, in systems of few quantum particles, a Schrodinger solution of the electronic wave function to be self-consistently coupled to the Poisson equation solution. For indirect bandgap materials such

as Si, care must also be taken to properly describe the multi-valley physics arising from confined electrons at low temperatures. A variety of software frameworks offer varying levels of these capabilities.

In addition to modeling pristine devices, the semiconductor industry also relies on a variety of tools that model the *processes* that form the devices, and can describe a variety of nonideal conditions such as charged defects, interface miscut, strain, and other effects. Much of the success of TCAD modeling in the semiconductor industry comes from the tight coupling of process and device simulation tools that allows the defects coming from various process steps to be empirically parameterized and then described as a computational model in device simulations. Analogous capabilities in the world of quantum devices are either missing or incomplete. In classical semiconductor devices, defects can be treated statistically. This statistical treatment of defects is not appropriate in the quantum regime, making both their experimental characterization and their inclusion into computational models more difficult. Consider the conduction of electrons through a narrow constriction: in a classical device, the current can be described by a perturbation arising from the statistical behavior of the defects in the constriction, whereas in a quantum device, the scales and the defect levels are often such that a device either has no defects, or has several defects, and the behavior of the two will be substantially different. Despite the difficulty of characterization and parameterization of the defect types and levels in quantum devices, such a capability promises substantial improvements in our ability to design robust semiconducting quantum devices. Currently a variety of nanofabrication techniques are used, including standard optical lithography, lift-off techniques, e-beam lithography, and even lithography using scanning-tunneling

microscope tips. Each technique imparts different defects on a device, and designers of qubit structures need to be able to consider these defect levels as part of their designs.

Because of their composite nature, superconducting qubits are more resistant to static defects such as fixed charge inclusion in devices. However, the solution of the equations governing superconductivity is more difficult, and typically requires a “full-field” solution of Maxwell’s equations, rather than the electrostatic description that suffices for semiconducting devices. Moreover, the superconducting quantum device field cannot fully leverage the investment in tools that the classical semiconductor industry has made. Full-scale simulations of superconducting quantum computing circuits are made with conventional microwave modeling packages that employ full vector finite element models. These models include the impact of dielectric constant, but must approximate superconductivity as metals with a priori defined resistivity of zero. Therefore, current simulations of superconducting quantum circuits overestimate loss and decoherence mechanisms and neglect quantum interactions and the details associated with superconductivity near surfaces and interfaces.

In many ways, trapped ion quantum devices have significant advantages over both semiconducting and superconducting quantum devices. The development of surface traps allows semiconductor fabrication techniques to be used to design small-scale, reproducible electrostatic traps, and the fact that the ions forming the qubits float above the surface traps make them unaffected by many of the semiconductor defects. However, the performance of trapped ion quantum devices is limited by the anomalous ion heating arising from imperfections in the fields produced by these

traps, which is believed to result from contaminants and transient inhomogeneity on the surface of the traps themselves. Understanding the sources of ion heating, to enable device designs that either reduce the rate of surface contaminants or perform in such a way as to be robust to such formation, will improve the performance and the reproducibility of trapped ion quantum devices.

### **Potential Impact**

Development of a suite of modeling tools that enable accurate design of quantum circuits using integrated predictive modeling to connect fabrication to materials to device and system performance, in analogy to today’s CMOS design and fabrication workflows, would be revolutionary. Accurate models calibrated to individual quantum devices will allow operating voltage ranges and interaction parameters to be largely determined theoretically, dramatically reducing the long tune-up times, allowing erroneous structures to be rapidly identified and eliminated, and yielding a tighter connection to pulse engineering and dynamical gate correction approaches that can improve and even optimize device performance. Moreover, such a capability allows optimization of device layouts to minimize crosstalk and noise correlations and improve entanglement. Beyond the quantum computing world, many of these issues also confront traditional semiconducting microelectronics, such as beyond-CMOS and spintronics devices, which will also be informed and accelerated by a fundamental knowledge of the device physics at the nano and quantum levels.



## **Eliminating Deleterious Environmental Effects Through Advanced Materials Manipulation**

The global environment in which a qubit resides is equally important to qubit performance as the local materials from which the qubit is constructed.

Improvements in the surrounding equipment necessary to conduct experiments in quantum computing can dramatically reduce current difficulties related to experimental research by better separating extrinsic and intrinsic effects.

### ***Opportunity***

Superconducting quantum computing is extremely sensitive to thermal energy entering the device because it results in electrons (a quasiparticle population in an otherwise perfectly conducting superconductor) that directly impact Cooper pair lifetimes. In addition to quasiparticles, residual magnetic fields cause losses in superconducting devices. To mitigate these loss mechanisms, non-magnetic connectors and components are being developed that are significantly less magnetic than those required in modern day nuclear magnetic resonance imaging devices used for medical applications.

For ion trap quantum computing, the electronic states of an ion that are used to execute coherent quantum operations require laser wavelengths near the transition frequencies separating the states. These wavelengths are typically in the blue or near-ultraviolet regions of the optical spectrum, and consequently optical materials for blue/UV laser light generation and delivery and blue/UV ion fluorescence detection are needed.

### ***Timeliness***

Directed effort toward improving filtering, fiber delivery, and shielding would enable accelerated advances in the experimental implementation of quantum computing technologies, allowing researchers to focus on the fundamental scientific issues underpinning these technologies.

### ***Approach***

For quantum computing devices operated at cryogenic temperatures, advances in shielding and filtering approaches, understanding the mechanical strains resulting from differential thermal expansion of materials cooled to millikelvin temperatures, and thermal and electrical performance at millikelvin temperatures are all valuable areas of study that can enable quantum computing research.

For ion trap technologies, narrow-band and tunable radiation sources (frequency converted to reach the necessary wavelength), bulk optical components, and fluorescence detectors (all of which reside outside the vacuum system) are used to generate laser light, point, and focus this light to a beam waist size of  $<10\text{-}\mu\text{m}$  diameter (and as small as  $\sim 2\text{ }\mu\text{m}$ ) at the ion, and then collect the laser induced fluorescence from the ion. While bulk external optics, sources, and detectors may work for small numbers of ions, significant increases in the number of ion qubits will require many more laser beams for the necessary optical pumping, laser cooling, and qubit manipulations, and many more optical transmission devices and detectors.

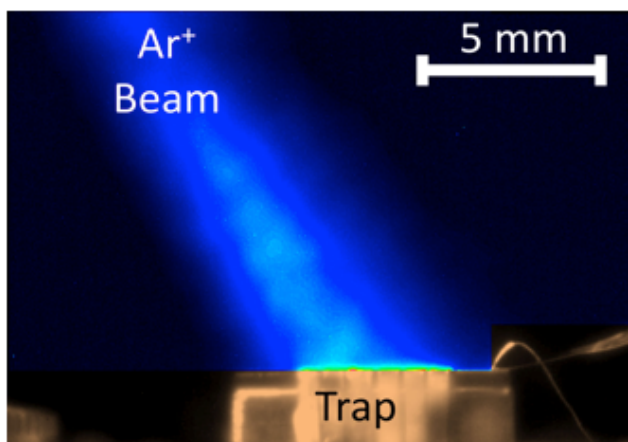
Fiber optics capable of delivering high power beams of up to tens of mW at wavelengths from 200-400 nm will be necessary, whether the pointing and focusing optics are outside the vacuum chamber in the form of bulk

optics or an array of MEMS mirrors or in close proximity to the ion at the trap chip in vacuum such as waveguides or diffractive optic elements. Minimal beam-pointing fluctuations and well-defined single mode profiles for the fibers will be necessary, as will minimized and stable attenuation characteristics and in some cases polarization maintenance. Hydrogen loading and fluorine doping of glass allow for near term improvements, while hollow core fibers offer the potential of significant power and reliability breakthroughs in the long term. Additional materials research, both theoretical and experimental, is needed for understanding and mitigating the basic mechanisms causing solarization in UV optical materials. Also, materials are needed for chip-scale waveguide routing of UV light, perhaps using nanophotonic phased arrays.

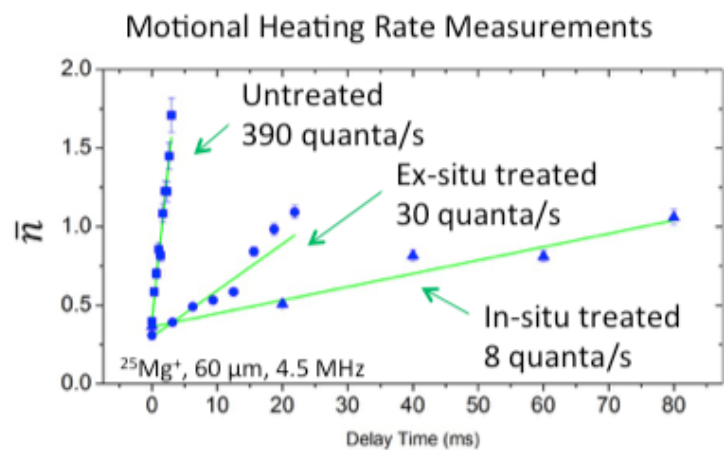
UV solid-state and semiconductor diode lasers that are tunable and narrow-band at the multiple necessary UV wavelengths are needed with improved frequency and power stability, as ion qubit gate fidelities critically depend on these two factors. In some instances, semiconductor lasers can currently

attain up to a few hundred mW at short wavelengths; however, narrower line widths, increased power, and shorter wavelengths are needed. Improvements in nitride semiconductor materials may lead to higher performance and shorter wavelength UV laser diodes. Also, frequency converted lasers, particularly fiber lasers, are candidates for attaining the requisite high powers at short wavelengths; however, materials improvements are essential to make these systems more robust, compact, and efficient.

Measuring the presence or absence of laser-induced fluorescence from the ion and counting the emitted photons determine the ion qubit state. Improved speed and efficiency are needed for the detectors counting these photons, as are improvements in the collection optics delivering the fluorescence to the detector. Both the fluorescence delivery optics and the detectors are currently outside of the vacuum chamber. Arrayed detectors, such as superconducting nanowire single-photon detectors, whose quantum efficiencies, speed, and latencies are optimized for specific UV



*Ar<sup>+</sup> plasma cleaning the electrode surfaces has shown to considerably decrease the anomalous heating rates of ion traps. (McKay 2014)*



wavelengths, seem to present a rich materials research opportunity, particularly within the context of integration of these detectors with surface electrode ion trap chips.

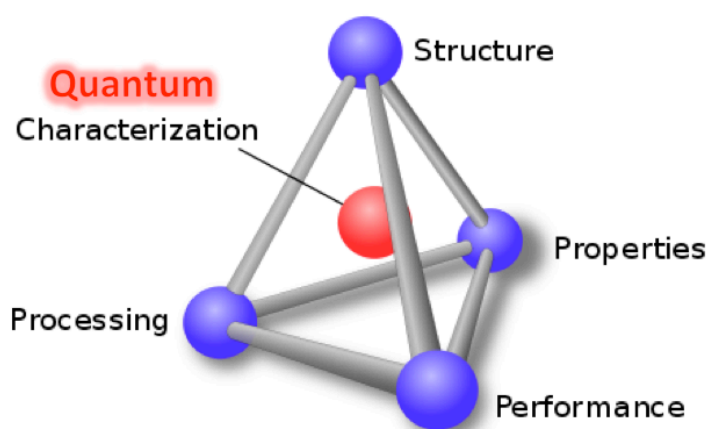
Much current research using trapped ions for quantum information processing employ surface electrode ion traps in which the ion qubit is trapped using RF and DC electric fields at distances of  $<100\text{ }\mu\text{m}$  above the electrode surface. As the ion is particularly sensitive to electric field noise, efforts are taken to clean all surfaces in proximity to the ion and to minimize or eliminate any unpinned electric potentials. Since dielectric materials represent a potential source of unwanted charge near the ion, electrically conductive, UV-transparent coatings are also needed for UV optical materials near the ion.

### ***Potential Impact***

Eliminating deleterious environmental effects through advanced materials manipulation will enable the more rapid experimental implementation of these quantum computing technologies and enable focus on the underpinning scientific issues that form the critical barriers to qubit performance. The UV light collection at the National Ignition Facility and low noise cryogenic measurements will also directly benefit from materials research efforts of supporting components needed for quantum computing experiments.

## Path Forward

The challenges associated with loss and decoherence from both static and dynamic mechanisms represent barriers for qubit performance and embody challenges at the frontiers of materials research. The transition of moving from electronic-grade to quantum-grade materials has implications far beyond the performance of individual qubit technologies. Moreover, to effectively exploit qubits for computation and not just demonstration, their performance must be consistent and reproducible, necessitating systematic approaches to characterization and fabrication grounded in models as well as databases and information repositories for the resulting insights.



*A balanced interdisciplinary approach is expected to produce the greatest impact of advancing materials science related to quantum computing.*

Importantly, the challenge extends beyond the qubit to the environment in which it resides. To visualize this challenge, consider a deconstructed qubit in which the device resides in a cryogenic measurement apparatus. Cryogenic measurement is critical

to reduce the energy of the entire system to levels that are significantly below the energy splitting of the qubit's two levels. The effects of the external equipment must be shielded, and this typically requires significant electrical, magnetic, and optical shielding. Within the sample space, harmful fluctuators must be removed. Inside the qubit material itself, the noise dynamically evolves based on charge and magnetic flux fluctuations on a variety of time scales from femtoseconds to seconds depending on the chemical composition of the material and the specific instantiation of impurities and disorder as well as its coupling to the measurement environment. By nature, qubits are sensitive to the periodic, amorphous, or disordered arrangement of atoms in the bulk and at interfaces.

Although the connection between materials properties and qubit performance is clear, today's design strategies are largely empirical. To move from Edisonian, trial-and-error approaches, to predictive design, a tightly coupled measurement-modeling-synthesis loop is required. The "quantum"-ness of the problem sets a high bar relative to materials by design strategies.

Given the magnitude of the challenge as well as the progress to date in the field, integrated team efforts with expertise that span quantum computing and materials research would seem to hold great promise for success. Heroic efforts by single investigators and small groups have laid a strong foundation; the opportunity is to systematize and accelerate these gains. In so doing, there is also the opportunity to grow a generation of interdisciplinary researchers equally fluent in qubit physics and materials research approaches.

Much of the needed materials research capability to meet the challenge of qubit technology is resident at the DOE laboratories, including especially their suite of national user facilities. Further, the breadth of the problems to be addressed demands multiple capabilities from multiple institutions.

As summarized in the U.S. Department of Energy Basic Energy Sciences report on mesoscale science<sup>21</sup>, “The requirement of quantum computing to minimize interaction with external degrees of freedom is counter to the usual goal of mesoscale science, which is to maximize the interactions among multiple degrees of freedom to promote emergent behavior. Although opposite in outcome, the two objectives rest on a common foundation: experimentally observing, theoretically understanding, and functionally controlling the interaction of disparate degrees of freedom. Whether the goal is to promote or inhibit, a firm observational base and predictive understanding of the interacting degrees of freedom are essential.”

As such, developing coordinated networks of researchers that span multiple laboratories to focus on these challenges in a systematic way holds great promise. Bringing the collective capabilities of the DOE national laboratory system to bear on these problem leverages billions of dollars of government investment.

The workshop that this report summarizes has already identified “low-hanging fruit” in which spontaneous collaborations bringing together existing expertise can make significant progress on known problems. The energy and enthusiasm workshop attendees brought to these discussions is a testament to the exciting opportunities that exist. An

ongoing series of workshops that highlights not only the successes but also the “failures” of these nascent collaborations has the potential to further expand these synergistic interactions.

Additional progress would be facilitated by a systematic and well-documented determination of the current limits of structural and chemical disorder in fabricated qubit devices by applying the most advanced currently existing materials characterization techniques and instruments to the problem. Additionally, materials by design and continued modeling of quantum materials may identify new synthetic techniques and material systems for quantum computing. In this pursuit the identification of model systems and platforms, including the determination of when “good” is good enough, is essential.

As expertise is systematically developed, distributed crosscutting centers focusing on key material areas such as atomic order/disorder, surfaces and interfaces, and multiscale phenomena with respect to the four canonical quantum computing technologies (including both existing tool utilization and development of new capabilities) are needed.

Finally, investments in capital improvements to advance the technical limits of existing capabilities are needed. This includes both the opportunity to invent and discover new tools as well as to expand the measurement environments and domains of applicability to existing capabilities. As the suite of key capabilities for enhancing qubit performance becomes clearer, the creation of community “test bed” facilities for systematic validation will also be invaluable.

## Conclusion

This is an exciting time at the nexus of quantum computing and materials research. The materials frontiers described in this report represent a significant advance in electronic materials and our understanding of the interactions between the local material and a manufactured quantum state. Simultaneously, directed efforts to solve materials issues related to quantum computing provide an opportunity to control and probe the fundamental arrangement of matter that will impact all electronic materials.

An opportunity exists to extend our understanding of materials functionality from electronic-grade to quantum-grade by achieving a predictive understanding of noise and decoherence in qubits and their origins in materials defects and environmental coupling. Realizing this vision systematically and predictively will be transformative for quantum computing and will represent a qualitative step forward in materials prediction and control.

This report attempts to capture the enthusiasm and energy of workshop participants not only in recognizing the grand challenge of materials advances required to enable quantum computing but also in charting a roadmap to seize the opportunity. Three grand challenges were identified that will provide the building blocks for success:

- Mechanisms that drive fundamental physics interactions across scales
- Engineered materials with exquisite control of electronic and physical structure, and
- Techniques to tease out novel properties of materials at unprecedented resolution.

Additionally, nine specific priority research directions were identified that embody the

grand challenges and provide an agenda for advancing materials research and empowering quantum computing:

- Sensing the local quantum environment through “qubit spectroscopy”
- Predicting microscopic origins of qubit material interactions
- Controlling atomic order exquisitely
- Imaging qubit materials in real time and multiple dimensions
- Connecting atomic fundamentals to qubits through mesoscale modeling
- Revealing qubit dynamics and decoherence mechanisms
- Cultivating the precise fabrication of ultrapure qubit devices
- Authenticating qubit devices using integrated models
- Eliminating deleterious environmental effects through advanced materials manipulation

Success in meeting these challenges will require a concerted effort of teams of researchers spanning quantum computing and materials research with expertise in theory and simulation, structural and physical characterization, and materials synthesis and fabrication. Further, while near-term opportunities exist to leverage existing capability, investment in the discovery and invention of new methods and techniques will be required for success. Finally, funding agencies will need to enhance and maintain world-leading capability in multiscale materials simulation, characterization capabilities with state-of-the-art resolution and sensitivity, and synthesis of material structures with exquisite compositional and geometrical control. Such investments will transform the role of materials science related to quantum computing and truly empower a new age of computation.



## Appendix 1: Materials Opportunities for Quantum Computing Workshop Agenda

**October 7-8, 2014**

**Los Alamos National Laboratory**

**WORKSHOP OBJECTIVE:** The goal of this workshop is to provide a forum for quantum device physicists to openly engage experts in broader areas of materials characterization and computational simulations in order to identify areas of potential research. This is a collaborative workshop between various Department of Energy laboratories and the Laboratory for Physical Sciences. The deliverable of this workshop will be a report that is circulated to the Department of Energy that can be used to motivate funding in materials science topics related to quantum computing. To foster a robust discussion at this workshop, technical presenters and breakout section leaders are expected to represent a broad perspective of their areas of expertise including: introductory material, results that represent the state of the art, current capabilities, and future prospects.

**Tuesday OCTOBER 7**

**All plenary sessions will be in Los Alamos Research Park Room 203A/B**

### **Plenary Session (Session Chair: Sarrao/Taylor, LANL)**

08:00-08:30	Registration/Logistics - Taylor/Sarrao (LANL)
08:30-08:45	Welcome and Workshop Goals - Richardson (LPS)
08:45-09:15	Quantum Computing - Lopata (LPS)
09:15-09:45	Semiconductor Donor Qubits – Lyon (Princeton)
09:45-10:15	SiGe Quantum Dots – Eriksson (Wisconsin)
10:15-10:45	BREAK
10:45-11:15	Enabling Technologies for Ion Traps Experiments - Leibfried (NIST)
11:15-11:45	Surface States in Ion Traps - Hite (NIST)
11:45-12:15	Superconductor Qubits - Pappas (NIST)
12:15-13:30	LUNCH (Research Park Room 203 A/B)

### **Quantum Materials Simulation Frontiers**

**Chair: E. Schwegler, LLNL**

13:30-13:45	Simulation Talk: Device Physics, D. Smith, LANL
13:45-14:00	Simulation Talk: Noise/Interfaces, V. Lordi, LLNL
14:00-14:15	Simulation Talk: ASCR Effort, N. Baker, PNNL
14:15-14:30	Simulation Talk: Semiconductors, R. Muller, SNL
14:30-14:45	General Discussion/Q&A – All Talks
14:45-15:00	BREAK

15:00-16:30 Breakout Sessions: Quantum Materials Simulation Frontiers  
**Locations and topics on page 61**

16:30-17:30 Reports from Breakout Groups  
(Muller, SNL; DuBois, LLNL; Stocks, ORNL; Bruillard, PNNL)

### **Wednesday OCTOBER 8**

**All plenary sessions will be in Los Alamos Research Park Room 203A/B**

08:00-08:15 Summary of Issues from Previous Day/Plan for the Day –Richardson, LPS

### **Quantum Materials Synthesis/Characterization Frontiers**

**Chair: L. Terminello, PNNL**

08:15-08:30 Syn/Char Talk: Spin Noise Spectroscopy, S. Crooker, LANL

08:30-08:45 Syn/Char Talk: Thin Film/Superconductor Fab, Q. Jia, LANL

08:45-09:00 Syn/Char Talk: X-ray Microscopy/Photon Sources, G. Ice, ORNL

09:00-09:15 Syn/Char Talk: Neutron Reflectometry, R. McQueeney, ORNL

09:15-09:30 Syn/Char Talk: Optics, Dawson, LLNL

09:30-09:50 Syn/Char Talk: Scanned Probe Microscopy, Ezra Bussmann, SNL,  
& Ilke Arslan, PNNL

09:50-10:05 General Discussion/Q&A – all talks

10:05-10:20 Break

10:20-12:00 Breakout Sessions  
**Locations and topics on page 61**

12:00-13:00 LUNCH (Research Park Room 203 A/B)

13:00-14:00 Reports from Breakout Groups  
(Taylor, LANL; Baddorf, ORNL; Chisolm, ORNL; Warner, PNNL)

14:00-15:00 Panel Discussion 1: QC Technologies Perspective  
(Somma, LANL; Carroll, SNL; Pappas, NIST; Leibfried, NIST; Hite, NIST;  
Lyon, Princeton)

15:00-16:00 Panel Discussion 2: Materials Science Perspective  
(Civale, LANL; Terminello, PNNL; Foiles, SNL; Allard, ORNL)

16:00-17:00 Wrap Up/Summary/What Did We Miss

Adjourn

## **Locations and Topics for Breakout Sessions**

### **Tuesday 15:00 – 16:30**

#### **Breakout Sessions: Quantum Materials Simulations Frontiers**

- #S1: Simulation of Quantum Dots and Ion Trap Qubits; Lead: Rick Muller, SNL  
— Research Park, Camelina Suite 200 (19)
- #S2: Simulation of Solid-State Qubits; Lead: Jonathan DuBois, LLNL  
— JRO Collaboration Space, JRO1 (30)
- #S3: Theory and Simulations of Qubit Materials; Lead: Malcolm Stocks, ORNL  
— JRO Collaboration Space, JRO2 (15)
- #S4: Simulation of Noise and Disorder; Lead: Paul Bruillard, PNNL  
— Research Park, Cassava Room 301 (16)

### **Wednesday 10:20 – 12:00**

#### **Breakout Sessions: Quantum Materials Synthesis/Characterization Frontiers**

- #M1: Optics and Photonics for Quantum Materials; Lead: Toni Taylor, LANL  
— JRO Collaboration Space, Retro Room (15)
- #M2: Superconducting Materials Fabrication & Synthesis; Lead: Matt Chisolm, ORNL  
— Research Park, Cassava Room 301 (16)
- #M3: Semiconducting Materials Fabrication & Synthesis; Lead: Art Baddorf, ORNL  
— Research Park, Camelina Room 200 (19)
- #M4: Imaging of Quantum Materials; Lead: Marvin Warner, PNNL  
— Research Park Conference Center Room 203A/B (60)

## Appendix 2: Workshop Participants

### ***Hosts***

Toni Taylor  
John Sarrao  
Brian Bluhm

Leonardo Civalé  
Quanxi Jia  
Darryl Smith  
Jian-Xin Zhu

Bruce Arey  
Ilike Arslan  
David Willingham  
James Evans

### ***Report Committee Members***

Toni Taylor  
John Sarrao  
Chris Richardson  
Paul Lopata  
Vince Lordi  
Jonathan DuBois  
Eric Schwegler  
Jay Dawson  
Matt Horsley  
Rebecca Nikolic  
Steve Libby  
Nathan Baker  
Marvin Warner  
Malcolm Carroll  
Rick Muller

Dmitry Yarotski  
Rolando Somma  
Terry Holesinger  
Vince Lordi  
Jonathan DuBois  
Eric Schwegler  
Jay Dawson  
Jeff Bude  
Matt Horsley  
Rebecca Nikolic  
Art Nelson  
Everett Wheelock  
Dave Knapp  
Steve Libby  
Gene Ice  
Kelly Perry  
Alan Tennant

Paul Bruillard  
Paul Clem  
Mike Sinclair  
Matt Blaine  
Malcolm Carroll  
Miek Wanke  
Greg Ten Eyke  
Joe Simonson  
Rick Muller  
Chris DeRose  
Nancy Missert  
Mary Crawford  
Chris Richardson  
Michael Mandelberg  
Vince Ballarotto  
Brad Blakestad  
Ben Palmer

### ***Participants***

Toni Taylor  
John Sarrao  
Scott Crooker

Larry Allard  
Chris Arrdahl  
Nathan Baker  
Lou Terminello  
Marvin Warner

Kevin Osborn  
David Moehring  
Marvin Kruger  
Chris Mineo  
Paul Lopata

## Figure Credits

C. J. K. Richardson, "Opening Remarks," *Materials Opportunities for Quantum Computing Workshop*, Los Alamos National Laboratory, October (2014).

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Publishing services were provided by Los Alamos National Laboratory.  
The co-chairs would like to thank Karen Kippen, Jim Cruz, and Diana Del Mauro  
for their support in editing, designing, and producing this report.

